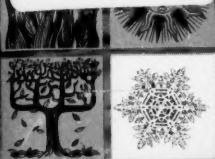1970
Quarterly
Technical
Progress
Review
Volume 13 – 4

A publication of the
Division of Technical Information
U.S. Atomic Energy Commission
Prepared in Cooperation
with
The American Nuclear Society.

REACTOR TECHNOLOGY

TECHNICAL PROGRESS REVIEWS

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Nuclear Safety
Wm. B. Cottrell, W. H. Jordan, J. P. Blakely, and associates
Nuclear Safety Information Center, Oak Ridge National Laboratory

Reactor Technology
Prepared in cooperation with the American Nuclear Society

All are available from the U. S. Government Printing Office. See the back cover for ordering instructions.

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United States Atomic Energy Commission (USAEC) reports are available at certain libraries that maintain collections of these reports. The libraries are listed on the inside front cover of each issue of *Nuclear Science Abstracts*. USAEC reports are also sold by the following governmental and international organizations: (1) National Technical Information Service (NTIS), U. S. Department of Commerce, Springfield, Va. 22151 (the price per copy of most separately bound documents is $3.00 for reproduced copy or $0.95 for microfiche); (2) International Atomic Energy Agency (IAEA), Vienna, Austria; and (3) National Lending Library, Boston Spa, England.

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REACTOR
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(This issue is arbitrarily designated Winter instead of Fall to coincide with the time of publication.)

COVER: Rapsodie Reactor at Centre
d'Études Nucléaires de Cadarache (France).
Photograph by Loïc-Jahan, Marseille.
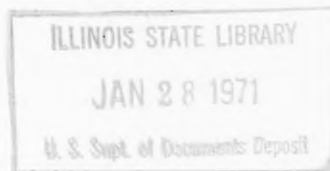
REACTOR TECHNOLOGY

is a quarterly review of progress and developments in the reactor field including the following subject areas:

Economics	Fuel Cycles
Physics	Fluid and Thermal Technology
Mechanics	Fuel Processing
Construction	Components
Fuel Elements	Operating Performance

Prepared for reactor physicists, reactor operators, designers, fuel-cycle specialists, administrators, and others interested in the field, this journal reviews, summarizes, critically evaluates and interprets the state of the art as reflected in the current literature.

The many reports and publications referenced merit further study, and readers are urged to consult them for additional details.

Qualified authors are invited to contribute review articles for consideration for publication.

Sodium Piping for the LMFBR

By E. M. Simons*

In 1968 the AEC published a 10-volume program plan (Reports WASH-1101–1110) for the development of a safe, reliable liquid-metal-cooled fast breeder reactor (LMFBR). Included in this prodigious work is a section delineating the technology required for the design and fabrication of coolant transport and storage systems and spelling out a plan for developing this technology.[1] As part of the implementation of this plan, C. F. Braun and Company, with the assistance of United Nuclear Corp., prepared a "Design Guide for LMFBR Sodium Piping," which is in the process of being published in interim form.

The LMFBR piping-design guide is based on currently available technology of high-temperature piping and experience with sodium systems. In areas where there is not enough background to permit LMFBR piping systems to be designed with the necessary assurances of safety, conservative approaches are recommended. Meanwhile, in accordance with the program plan, studies are being conducted to fill in the gaps. It is planned to utilize the findings as they become available to update the guide.

Actually the severe combination of design conditions for a 1000-MW(e) LMFBR sodium-piping system requires engineering considerations beyond the scope of normal handbook design. The largest piping ever used for sodium service in the United States is 30-in.-diameter, $\frac{3}{8}$-in.-wall 304 stainless-steel pipe carrying 800°F (427°C) sodium in 70-ft runs from the reactor vessel to each of the three intermediate heat exchangers in the Enrico Fermi Fast Breeder Reactor (EFFBR). Thin-walled austenitic stainless-steel piping, 12 in. in diameter or smaller, has been used extensively

in all sodium-cooled reactors operated to date. However, the individual designers had to make their own modifications of existing codes to ensure the integrity of the piping under the high temperatures, thermal transients, and difficult maintenance conditions encountered in nuclear sodium service.

Conceptual designs of 1000-MW(e) LMFBRs call for primary-system sodium piping up to 48 in. in diameter and maximum sodium temperatures in the neighborhood of 1200°F (650°C). This article presents considerations regarding sodium-piping technology in relation to the requirements of full-scale LMFBR systems. Included are some highlights of the proposed guide.

SYSTEM REQUIREMENTS

Steady-state operating conditions were defined for various 1000-MW(e) LMFBRs proposed in conceptual designs by five different AEC contractors. These conditions were used by C. F. Braun and Company[2] to define the range of normal operating parameters for use in preparing the guide. They are shown in Table 1.

CODE DESIGN CRITERIA

In August 1969 the USA Standards Institute approved[3] the Nuclear Power Piping Code USAS B31.7. This standard provides requirements for the design, materials, fabrication, assembly, erection, installation, and examination of three classes of piping systems for nuclear power-plant service. It parallels Section III, Nuclear Vessels, of the *ASME Boiler and Pressure Vessel Code*, reflecting the similarities in requirements for nuclear vessels and the associated piping.

These codes are being used to ensure the safe design of vessels and piping for water-cooled nuclear

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Table 1   Range of LMFBR Steady-State Operating Conditions[2]

Parameter	Primary system		Secondary system	
	Maximum value	Minimum value	Maximum value	Minimum value
Plant life, year	30	30	30	30
Sodium temp., °F				
Hot leg	1200	900	1165	800
Cold leg	970	600	850	500
Loop ΔT	300	230	315	250
Maximum pressure				
Internal, psig	175	70	90	90
External, psig	20	0	0	0
Loop ΔP, psi	155	45	90	30
Number of loops	6	2	4	2
Sodium flow rate				
Total, 10^6 lb/hr	127.4	16	106.5	92
Per loop, 10^6 lb/hr	63.7	5.1	46	5.0
Per loop, 10^3 gal/min	149.2	12	110	12
Pipe size				
Hot leg, in.	48	24	44	16
Cold leg, in.	42	18	42	16
Cover gas				
Kind	He or Ar	He or Ar	He or Ar	He or Ar
Purity, %	99.997	99.8975	99.997	99.8975
Pressure, psi	30	3 in. H_2O	100	3 in. H_2O
Radioactivity				
Sodium, mR/hr	3.8×10^8	9.1×10^7	1	0.25
Outside pipe, mR/hr	3.8×10^8		< 0.2	
Impurities in sodium				
Oxygen, ppM	10	<10 (sat'n <325°F)	10	
Hydrogen, ppM	Sat'n <325°F	0.002		
Carbon, ppM	20	20		

power plants of the type being built today. As adopted, they limited the temperatures to 700°F for carbon and low-alloy steels and 800°F for austenitic steels. Subsequently Code Case Interpretation 1331 was issued permitting the use of code rules, with modifications, up to 1100 and 1200°F, respectively.

On the basis of a pointed study,[4] C. F. Braun and Company and United Nuclear found that the existing nuclear piping and pressure-vessel codes are unsuitable for the design of LMFBR piping without modification. Some observations leading to this conclusion are:

1. Because of the very high thermal conductivity of sodium (20 times that of water), any rapid change in fluid temperature resulting from some transient condition will induce proportionally high thermal stresses in the pipe wall.

2. The target 30-year service life of the LMFBR dictates a lowering of the allowable high-temperature stress values found in the present nuclear codes (including Code Case 1331). These values are based on creep and rupture data extrapolated from relatively

short-term test results to 100,000 hr by techniques which, although apparently conservative, cannot be employed with assured safety for further extrapolation to the 200,000-hr values needed for the LMFBR.

3. No distinction is made in USAS B31.7 between short-term (transient) and long-term (from differential thermal expansions) thermal stresses; both are considered as self-limiting secondary stresses that also contribute to localized peak stresses. The "stress-range" concept employed by the code for cyclic secondary stresses permits some relaxation of stress during the hot portion of a thermal cycle—a hazardous assumption for short-duration radial thermal stresses.

It is planned to publish the guide in three volumes: I, Requirements; II, Procedures; and III, Stress Report. Volume I closely follows the format of USAS B31.7-1969, and some sections are identical. The major points of departure are related to the liquid-sodium coolant, the higher operating temperatures, and the

30-year service life under severe cyclic operating conditions. Considerable simplification was achieved by concentrating only on the LMFBR environment and on the very few candidate materials for LMFBR piping.

PIPING MATERIALS

Selection of Candidates

Much study has gone into the selection of structural materials for sodium systems. For example, in-depth evaluations have been made of properties, environmental effects, and weldability of candidate materials for FCR (Fast Ceramic Reactor),[5] FARET (Fast Reactor Test),[6] FFTF (Fast Flux Test Facility),[7] SPTF (Sodium Pump Test Facility),[8] and the various 1000-MW(e) LMFBR conceptual systems, to name a few.

In each case the foremost objective in the selection was to provide the utmost in system integrity, making maximum use of existing technology. Desirable characteristics of the piping materials for high-temperature sodium systems include:

1. Good strength, ductility, and resistance to creep and fatigue under the operating conditions.

2. Resistance to interaction with molten sodium at operating conditions.

3. Commercial availability (or potential) in the necessary sizes and fittings, with a well-developed technology.

4. Fabricability and weldability.

5. Air-side oxidation resistance and inertness to thermal insulation at operating temperatures.

6. High thermal conductivity and low thermal expansion to minimize thermal stress and fatigue problems.

In addition, any primary-system piping close to the reactor core must be able to resist deterioration by the anticipated fast-neutron fluence.

In the light of these desired properties, along with the conclusions from the aforementioned in-depth studies, three materials were selected as the basis for the guide: 304 and 316 stainless steels and 2.25 Cr−1 Mo steel. Other steels may eventually turn out to be superior to these (particularly with regard to carbon transfer). However, the materials selected are the most advanced commercially of those which meet most of the criteria. These materials are not only being used extensively in the nonnuclear utility and process industries but are also among the ones most frequently used in nuclear sodium systems to date (SRE, HNPF, EBR-II, EFFBR, SEFOR, LAMPRE-I, and Rapsodie).

Both 304 and 316 stainless steels are highly susceptible to stress-corrosion attack in aqueous media. Consequently 2.25 Cr−1 Mo steel was selected for the steam generators of a number of sodium-cooled reactor plants (HNPF, EBR-II, and EFFBR) and is the favored material for future LMFBR steam generators and possibly for the secondary-system piping. The cost of 2.25 Cr−1 Mo steel per pound is less than one-third of that of 304 stainless steel, which is a strong incentive for using it where its physical and mechanical properties are adequate. Higher chromium ferritic alloys have been deferred from consideration because of their higher cost and welding problems.

The intermediate heat exchanger in the LMFBR will most likely be made of 304 or 316 stainless steel, and the steam generator and part of the secondary-loop sodium piping will probably be 2.25 Cr−1 Mo steel. Experience has shown that such dissimilar materials in a polythermal sodium loop can give rise to mass-transfer (particularly carbon-migration) problems and difficulties with transition joints. The use of a stabilized grade of 2.25 Cr−1 Mo steel could mitigate the carbon-transfer problem, but such steels are not presently code accepted.

Materials Specifications

The composition and specifications tentatively recommended by C. F. Braun and Company and United Nuclear for the selected base and welding materials for LMFBR piping are presented in Tables 2 to 5. Note that only L (low carbon) and H (high carbon) grades of the stainless steels are included. The L grades should be used where there is danger of intergranular attack by aqueous media, because carbide precipitation (sensitization) is limited. On the other hand, since carbide precipitation does not seem to be harmful in sodium systems, it is probably desirable to take advantage of the better strength of the H grades for LMFBR sodium piping. Standard grades (no suffix) are not included in the tables because they carry only an upper limit for carbon (0.08 wt.%), and any low-carbon material used would not have the strength of the H grades.

The RDT materials standards listed in Tables 4 and 5 (see the first footnote in Table 4) specify the basic qualification tests that must be run, methods of removing and handling specimens, test conditions, procedures, and acceptance criteria for each material. Generally the tests include chemical analysis, short-time tensile tests, impact tests, and several nondestructive examinations. The design organization is free to

Table 2   Recommended Base-Material Compositions for LMFBR Piping

| | Grade | | | | | |
| | | | | | 2.25 Cr–1 Mo | |
Element	304L	304H	316L	316H	Ladle analysis	Check analysis
Carbon, %	0.035 max.	0.04–0.10	0.036 max.	0.04–0.10	0.15 max.	
Manganese, %	2.00 max.	2.00 max.	2.00 max.	2.00 max.	0.30–0.60	0.27–0.63
Phosphorus (max.), %	0.040	0.040	0.040	0.040	0.035	
Sulfur (max.), %	0.030	0.030	0.030	0.030	0.035	
Silicon, %	0.75 max.	0.75 max.	0.75 max.	0.75 max.	0.15–0.30	0.13–0.33
Nickel, %	8.00–13.0	8.00–11.0	11.0–15.0	11.0–14.0		
Chromium, %	18.0–20.0	18.0–20.0	16.0–18.0	16.0–18.0	2.00–2.50	1.88–2.62
Molybdenum, %			2.00–3.00	2.00–3.00	0.90–1.10	0.85–1.15

specify any added special material-qualification requirements that it considers necessary to ensure the integrity of the piping system.

Environmental Effects

A comprehensive review of the literature on corrosion and mass transfer of austenitic and ferritic steels in sodium systems was made by United Nuclear.[9] The purpose of this study was to ascertain how the strength properties of LMFBR piping materials would be affected by their 30-year exposure to sodium inside and to air or other environments outside.

Surface Dissolution. Solution attack, normally occurring in the hot leg of the system (between the reactor and the intermediate heat exchanger), leads to thinning of a pipe wall and consequent loss of load-carrying ability. To compensate for solution attack, the design guide recommends that the hot-leg pipe wall thickness shall be increased by a "corrosion allowance" obtained with the aid of Fig. 1 as the sum of all metal losses expected to occur during the estimated times at each predicted combination of temperature and oxygen content (5 ppM minimum) over the lifetime of the system. Finally a fixed 10-mil allowance (5 mils for 2.25 Cr–1 Mo steel) is added to the computed wall thickness to provide for some intergranular attack—a phenomenon that has been observed in only a few long-term laboratory loops and is poorly understood. If a pipe has sodium both inside and outside, as in pool-type primary systems, these corrosion allowances must be doubled.

Corrosion-Product Deposition. Mass-transfer deposits in the large-diameter cold-leg piping are expected neither to have much influence on heat transfer nor to cause excessive pressure drops. However, deposits of

activated corrosion products could necessitate additional shielding of the piping external to the reactor.

Carburization and Decarburization. In a mono-metallic sodium loop, carbon will migrate from the hot leg to the cold leg. In a bimetallic system composed of austenitic stainless steel and 2.25 Cr–1 Mo steel, the latter will be decarburized. Such carbon transfer can have significant effects on the properties of the pipe materials. In general, the weakening effect of decarburization is considered to be more serious than the embrittling effect of carburization.

The effects of carbon transfer are compensated for in the guide by "adjusting" the mechanical properties of the piping materials, using the multiplying factors shown in Table 6. Figures 2 and 3 permit the extent of decarburization after a 30-year service life to be estimated. As Table 6 shows, if the mean carbon level of the austenitic stainless steels is expected to fall below 0.04 wt.%, the L-grade properties should be used for piping design. Also, although some properties are actually enhanced by changes in carbon level, adjustment factors are limited to unity to be conservative.

It should be emphasized at this point that Table 6 is the result of scrupulous weighing of all available results from sodium–steel–carbon studies to date,[9] which are far from adequate for clear-cut application to piping-design needs. As current programs shed more light on the subject, the recommended piping-design procedures to compensate for carbon transfer will be modified accordingly.

Effects of External Environments. External environments of inert gas or oxygen-depleted air are not expected to influence the piping design. However, 316 stainless steel in a low-oxygen atmosphere must be kept below 1200°F to avoid catastrophic oxidation.

Table 3 Recommended Compositions of Welding Filler Materials for LMFBR Piping

Element and ASTM spec.*	AWS–ASTM classification									
	E308L†	E308†	E316L	E316	E16-8-2	E317	E9015-B3L (ASTM A-316)	E9015-B3L (ASTM A-316)	E9015-B3, E9016-B3 (ASTM A-316)	E9018-B3 (ASTM A-316)
Carbon, %										
A-298	0.04 max.	0.08 max.	0.04 max.	0.08 max.	0.10 max.	0.08 max.	0.05 max.	0.12 max.	0.12 max.
A-371	0.03 max.	0.08 max.	0.03 max.	0.08 max.					
Chromium, %										
A-298	18.0–21.0	18.0–21.0	17.0–20.0	17.0–20.0	14.5–16.5	18.0–21.0	2.00–2.50	2.00–2.50	2.00–2.50
A-371	19.5–22.0	19.5–22.0	18.0–20.0	18.0–20.0		18.5–20.5			
Nickel, %										
A-298	9.0–11.0	9.0–11.0	11.0–14.0	11.0–14.0	7.5–9.5	12.0–14.0			
A-371	9.0–11.0	9.0–11.0	11.0–14.0	11.0–14.0		13.0–15.0			
Molybdenum, %										
A-298			2.0–2.5	2.0–2.5	1.0–2.0	3.0–4.0	0.90–1.10	0.90–1.10	0.90–1.10
A-371			2.0–3.0	2.0–3.0		3.0–4.0			
Manganese, %										
A-298	2.5 max.	2.5 max.	2.5 max.	2.5 max.	2.5 max.	2.5 max.	0.90 max.	0.90 max.	0.90 max.
A-371	1.0–2.5	1.0–2.5	1.0–2.5	1.0–2.5		1.0–2.5			
Silicon, %										
A-298	0.90 max	0.90 max.	0.90 max.	0.90 max.	0.50 max.	0.90 max.	1.00 max.	0.60 max.	0.80 max.
A-371	0.25–0.60	0.25–0.60	0.25–0.60	0.25–0.60		0.25–0.60			
Phosphorus, %										
A-298	0.04 max.	0.04 max.	0.04 max.	0.04 max.	0.03 max.	0.04 max.	0.03 max.	0.03 max.	0.03 max.
A-371	0.03 max.	0.03 max.	0.03 max.	0.03 max.		0.03 max.			
Sulfur, %										
A-298	0.03 max.	0.03 max.	0.03 max.	0.03 max.	0.03 max.	0.03 max.	0.04 max.	0.04 max.	0.04 max.
A-371	0.03 max.	0.03 max.	0.03 max.	0.03 max.		0.03 max.			

*A-298 applies to covered welding electrodes; A-371 applies to welding rods and bare electrodes.
†Chromium, minimum = 1.9 x nickel, when so specified.

Table 4 Applicable Specifications for Base Materials for LMFBR Piping

| Grade designation | | | Specification | | |
AISI	ASTM	Description	ASTM	ASME	RDT*
304L or 316L	304L or 316L	Plate, sheet, strip	A-167		
		Pipe flanges, fittings, valves	A-182	SA182	M2-2 (MA182)
		Seamless tubing	A-213	SA213	M3-2 (MA213)
		Plate, sheet, strip	A-240	SA240	M5-1T (MA240)
		Welded tubing	A-249	SA249	
		Seamless tubing	A-269		
		Bars and shapes	A-276		M7-1 (MA276)
		Seamless and welded pipe	A-312	SA312	
		Welded pipe	A-358		M3-7
		Bars and shapes	A-479	SA479	M7-3T (MA479)
		Seamless tubing	A-511		
		Welded tubing	A-554		
304H or 316H	304H or 316H	Pipe flanges, fittings valves	A-182	SA182	M2-2 (MA182)
		Seamless tubing	A-213	SA213	M3-2 (MA213)
		Welded tubing†	A-249†	SA249†	
		Seamless and welded pipe	A-312	SA312	
		Welded pipe	A-358		M3-7
		Seamless pipe	A-376	SA376	M3-3T (M376)
		Forged and bored pipe	A-430	SA430	
2.25 Cr– 1 Mo	2.25 Cr– 1 Mo	Welded pipe	A-155		
	F22	Pipe flanges, fittings, valves	A-182	SA182	M2-2 (MA182)
	T22	Seamless tubing	A-199	SA199	
	T22	Seamless tubing	A-200		
	T22	Seamless tubing	A-213	SA213	M3-2 (MA213)
	P22	Seamless pipe	A-335	SA335	M3-12T (MA335)
	F22	Forgings	A-336	SA336	M2-4T (MA336)
	FP22	Forged and bored pipe	A-369	SA369	
	GrD‡	Alloy steel plate	A-387	SA387	M5-5T (MA387)

*Under the AEC Division of Reactor Development and Technology (RDT), the Oak Ridge National Laboratory (ORNL) is administering a comprehensive standards program under which RDT standards are issued for application on RDT programs. Under this program, standards that apply only to liquid-metal systems are being issued by the Liquid Metal Engineering Center. The numbers in parentheses are older designations.

†Applies to 304H only.

‡Annealed material only.

If the external environment is air, a corrosion allowance must be added to the wall thickness of 2.25 Cr–1 Mo steel in accordance with the following:

Temp., °F	Rate, mils/year
950 or below	1.0
1000	2.7
1050	5.2
1100	7.4

An air environment is not expected to have any detrimental effects on 304 or 316 stainless steel, except during shutdowns in the presence of certain harmful agents. To prevent such effects, the guide cautions against permitting certain types of materials to be left in contact with the piping for any length of time. These include (1) moisture; (2) a long list of common acids or acid-forming chemicals that combine with moisture and induce intergranular corrosion in austenitic stainless steels that have been sensitized by extended operation above 800°F; (3) halogen salts,

Table 5   Applicable Specifications for Welding Filler Materials for LMFBR Piping

| Grade designation | | | Specification | | |
AISI	ASTM	Description	ASTM	ASME	RDT*
304L	E308L	Covered welding electrodes	A-298	SA298	M1-1 (MA298)
	ER308L	Welding rods and bare electrodes	A-371	SA371	M1-2 (MA371)
304H	E308	Covered welding electrodes	A-298	SA298	M1-1 (MA298)
	ER308	Welding rods and bare electrodes	A-371	SA371	M1-2 (A371)
316L	E316L	Covered welding electrodes	A-298	SA298	M1-1 (MA298)
	ER316L	Welding rods and bare electrodes	A-371	SA371	M1-2 (MA371)
316H	E316	Covered welding electrodes	A-298	SA298	M1-1 (MA298)
	E16-8-2				
	ER316	Welding rods and bare electrodes	A-371	SA371	M1-2 (MA371)
2.25 Cr– 1 Mo	E9015-B3L	Covered welding electrodes	A-316	SA316	M1-4 (MA316)
	E9015-B3				
	E996-B3				
	E9018-B3				

*Under RDT, ORNL is administering a comprehensive standards program under which RDT standards are issued for application on RDT programs. Under this program, standards that apply only to liquid-metal systems are being issued by the Liquid Metal Engineering Center. The numbers in parentheses are older designations.

which, even in minute quantities, break down the passive film locally and cause pitting and crevice corrosion by electrochemical action; and (4) acid chlorides, fluorides, and caustic solutions that cause stress-corrosion cracking. Possible sources of such contaminants are cleaning agents, coatings on welding electrodes, insulating materials, and contaminants (including moisture) in the atmosphere. It is suggested that outdoor sodium piping be kept above the dew-point temperature at all times.

Radiation. In general, LMFBR piping will not be located where the neutron flux is unusually high. Consequently the piping designer will not have to worry about radiation embrittlement for the most part. However, if the anticipated neutron fluence at any section of piping exceeds 10^{18} neutrons/cm^2 (>1 MeV), the resulting loss of ductility of the piping in that region must be provided for in the design.

Physical Properties

Following a thorough evaluation of the widespread literature on basic properties of the selected materials,[9] C. F. Braun and Company and United Nuclear recommend the use of the values listed in Tables 7 and 8 for piping-design purposes.

Mechanical Properties

In similar fashion a set of mechanical properties of the selected materials in air was culled from the literature and recommended for use in the design of LMFBR piping. Typical data are shown in Table 9 and in Figs. 4 to 8. In many cases gross extrapolation or averaging of widely scattered data was required, and the design bases will remain questionable until more pointed results from long-term test programs now in progress become available.

The design stress intensities for sustained loading (S_{ms}) and for transient loading (S_{mt}) are given in Tables 10 and 11, respectively. All allowable stresses are keyed to these design values. The stresses were determined from the best available mechanical-property data at each temperature listed as the *lowest* of the following:

For both S_{ms} and S_{mt},

1. One-third of the specified minimum tensile strength at room temperature.

2. One-third of the tensile strength at temperature.

3. Two-thirds of the specified minimum yield strength at room temperature.

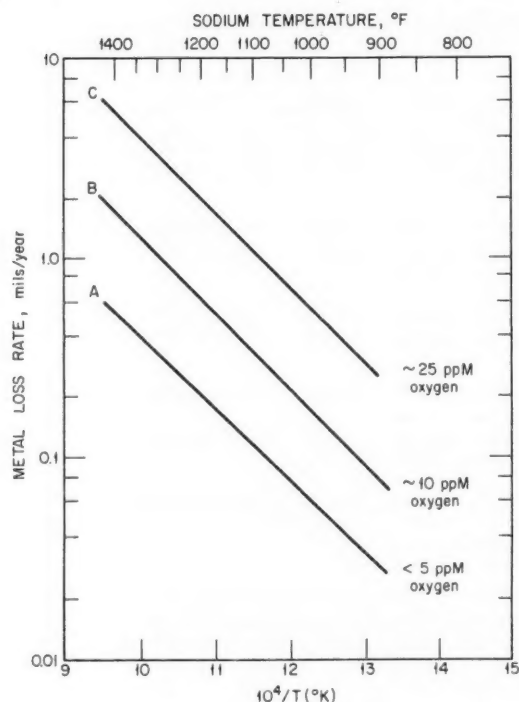4. Two-thirds of the yield strength at temperature (for austenitic stainless steel, 90% of the yield strength

Fig. 1 Anticipated rate of metal loss in hot-leg sodium piping.[9]

at temperature but not exceeding two-thirds of the room-temperature yield strength).

For S_{ms} only,

5. Two-thirds of the stress to produce rupture in 200,000 hr, based on average data extrapolated to 200,000 hr (value of average 200,000-hr rupture strength shall not exceed 125% of minimum value).

6. Four-fifths of the stress to produce rupture in 200,000 hr, based on minimum values.

7. One-hundred percent of the stress to produce a creep rate of 0.005% in 1000 hr.

To avoid confusion, the mechanical properties listed in the guide do not include any allowances for sodium corrosion effects. Adjustment factors and allowances for environmental effects must be applied separately, as discussed previously.

FABRICATION, INSTALLATION, AND TESTING

Recommendations in the guide for the assembly and testing of LMFBR piping are based largely on information in a comprehensive review by C. F. Braun and Company.[11] The requirements incorporate appropriate parts of USAS B31.7, Section III of the ASME code and the RDT specifications, along with all available experience with piping in previously designed sodium-cooled reactor plants.

Table 6   Multiplying Factors To Adjust Mechanical Properties of Piping Materials for Changes Caused by Prolonged Exposure to Sodium[9]

Material	Location	System type	Change	Mean C, wt.%	UTS*	YS†	SR‡	CR§	Fatigue	Impact
304H S.S. 304 S.S.	Hot leg	Monometallic	Decarburization	< 0.04	L¶	L	L	L	1.0	0.5
304H S.S. 304 S.S.	Hot leg	Bimetallic	Carburization	>0.10	1.0	1.0	1.0	0.65	1.0	0.5
316H S.S. 316 S.S.	Hot leg	Monometallic	Decarburization	<0.04	L	L	L	L	1.0	0.5
316H S.S. 316 S.S.	Hot leg	Bimetallic	Carburization	>0.10	1.0	1.0	1.0	1.0	1.0	0.5
2.25 Cr– 1 Mo	Hot and cold leg	Bimetallic	Decarburization	≤0.02	0.60	0.60	0.62	0.46	1.0	1.0
				0.03	0.68	0.68	0.62	0.52	1.0	1.0
				0.05	0.75	0.75	0.62	0.61	1.0	1.0
				0.07	0.83	0.83	0.62	0.72	1.0	1.0
				0.09	0.90	0.90	0.92	0.83	1.0	1.0
				0.11	0.97	0.97	0.92	0.94	1.0	1.0
				≥0.12	1.0	1.0	0.92	1.0	1.0	1.0

*UTS = ultimate tensile strength.
†YS = 0.2% offset yield strength.
‡SR = stress-to-rupture in 100,000 hr.

§CR = stress for secondary creep rate of 0.01% in 1000 hr.
¶L = use low-carbon-grade values.

Welding

Approved processes for welding LMFBR piping include tungsten–inert gas (TIG), consumable metal-arc inert gas (MIG), and submerged arc. Short-arc and oxyacetylene welding are not permitted. Pipe sections and attachments are to be joined with butt welds; socket welds or "stitch welds" are prohibited. Compo-

sitions and specifications for welding filler materials are shown in Tables 3 and 5, respectively.

Liquid sodium can attack welding slag or non-metallic inclusions in the welds. For this reason the root pass of any sodium-contacting pipe weld that is accessible only from the outside must be smooth, clean, free of defects, and should not be made by
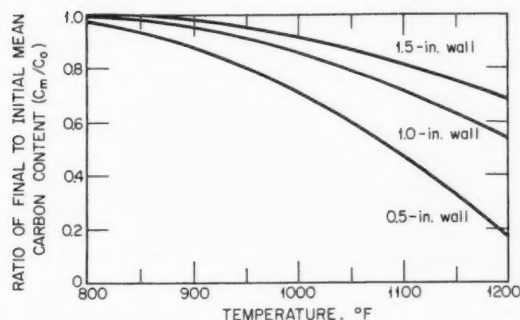
Fig. 2 Estimated mean carbon content in 304 and 316 stainless-steel monometallic system hot-leg sodium piping after 30 years.[9]
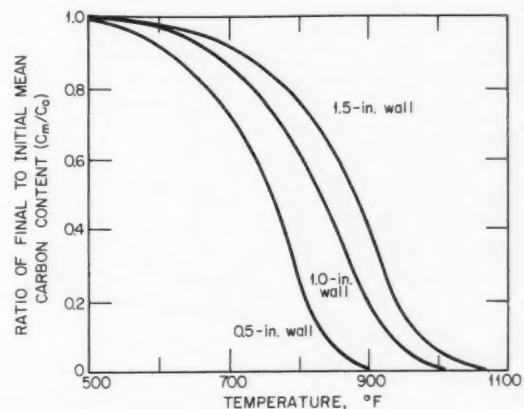
Fig. 3 Estimated mean carbon content in 2.25 Cr–1 Mo steel bimetallic system sodium piping after 30 years.[9]

Table 7 Values of Physical Properties of 304 and 316 Stainless Steels Recommended for Use in LMFBR Piping Design[9]

Temp., $^{\circ}$F	Young's modulus, 10^6 psi	Shear modulus, 10^6 psi	Poisson's ratio	Coefficient of thermal expansion		Thermal conductivity, Btu/(hr)(ft)($^{\circ}$F)	Specific heat, Btu/(lb)($^{\circ}$F)	Density,* lb/in.^{3}	Thermal diffusivity, ft^2/hr
				Mean above 70°F, $10^{-6}/^{\circ}$F	Instan-taneous, $10^{-6}/^{\circ}$F				
70	28.3	11.2	0.24	9.11	9.11	8.35	0.108	0.290	0.150
100	28.1	11.1	0.25	9.16	9.21	8.40	0.110	0.290	0.150
200	27.7	10.9	0.27	9.34	9.50	8.90	0.116	0.289	0.155
300	27.1	10.6	0.29	9.47	9.73	9.35	0.120	0.288	0.159
400	26.6	10.4	0.30	9.59	9.96	9.80	0.124	0.287	0.163
500	26.1	10.1	0.31	9.70	10.20	10.23	0.126	0.286	0.166
600	25.4	9.9	0.32	9.82	10.43	10.27	0.128	0.285	0.171
700	24.8	9.6	0.32	9.93	10.66	11.10	0.130	0.285	0.174
800	24.1	9.3	0.32	10.05	10.90	11.55	0.132	0.284	0.178
900	23.3	9.1	0.32	10.16	11.09	11.90	0.134	0.283	0.181
1000	22.5	8.8	0.31	10.29	11.28	12.42	0.137	0.282	0.185
1100	21.7	8.5	0.30	10.39	11.47	12.90	0.139	0.281	0.190
1200	20.9	8.2	0.29	10.48	11.65	13.30	0.141	0.280	0.193
1300	20.1	7.9	0.28	10.54	11.84	13.80	0.144	0.279	0.198
1400	19.2	7.6	0.27	10.60	12.01	14.20	0.147	0.278	0.202

*Values given are for 304 stainless steel; for 316, subtract 0.002.

Table 8 Values of Physical Properties of 2.25 Cr–1 Mo Steel Recommended for Use in LMFBR Piping Design

Temp., °F	Young's modulus, 10^6 psi	Shear modulus, 10^6 psi	Poisson's ratio	Coefficient of thermal expansion		Thermal conductivity, Btu/(hr)(ft)(°F)	Specific heat, Btu/(lb)(°F)	Density, lb/in.^{3}	Thermal diffusivity, ft^2/hr
				Mean above 70°F, 10^{-6}/°F	Instantaneous, 10^{-6}/°F				
70	29.9	12.0	0.26	6.07	6.07	16.25	0.107	0.282	0.421
100	29.8	11.9	0.27	6.13	6.20	16.25	0.108	0.282	0.417
200	29.5	11.8	0.28	6.38	6.67	16.25	0.111	0.281	0.408
300	29.0	11.6	0.29	6.60	7.10	16.25	0.115	0.281	0.393
400	28.6	11.3	0.30	6.82	7.54	16.25	0.119	0.280	0.382
500	28.0	11.0	0.30	7.02	7.96	16.25	0.124	0.280	0.366
600	27.4	10.7	0.30	7.23	8.35	16.25	0.131	0.279	0.348
700	26.6	10.4	0.30	7.44	8.76	16.25	0.138	0.278	0.331
800	26.0	10.1	0.30	7.65	9.16	16.25	0.145	0.277	0.317
900	25.3	9.8	0.29	7.84	9.52	16.25	0.154	0.277	0.298
1000	24.5	9.4	0.29	7.97	9.88	16.25	0.163	0.276	0.283
1100	23.7	9.0	0.28	8.12	10.20	16.25	0.172	0.275	0.269

Table 9 Design Yield Strength of LMFBR Piping Materials

Temp., °F	Yield strength, psi				
	304H	304L	316H	316L	2.25 Cr–1 Mo annealed†
75	30,000*	25,000*	30,000*	25,000*	30,000
100	28,800	24,250	29,100	24,250	30,000
200	24,900	21,250	25,800	21,250	28,000
300	22,500	19,250	23,400	19,000	26,900
400	20,700	17,500	21,000	17,500	26,200
500	19,500	16,250	19,800	16,000	25,800
600	18,300	15,500	18,900	15,250	25,000
650	18,000	15,250	18,450	14,875	24,300
700	17,700	15,000	18,000	14,500	23,500
750	17,250	14,750	17,850	14,125	22,800
800	16,800	14,500	17,700	13,750	22,400
850	16,500	14,250	17,550	13,500	12,700
900	16,200	14,000	17,400	13,250	21,100
950	15,900	13,625	17,250	12,875	20,500
1000	15,600	13,250	17,100	12,500	19,600
1050	15,150	12,875	16,800	12,125	
1100	14,700	12,500	16,500	11,750	
1150	14,400	11,875	16,350	11,125	
1200	14,100	11,250	16,200	10,500	
1250	13,650		15,750		
1300	13,200		15,300		
1350	12,450		14,850		
1400	11,700		14,400		

*Room-temperature yield points are specified minima; data at higher temperatures are normalized to these, using trend curves by Smith.[10]
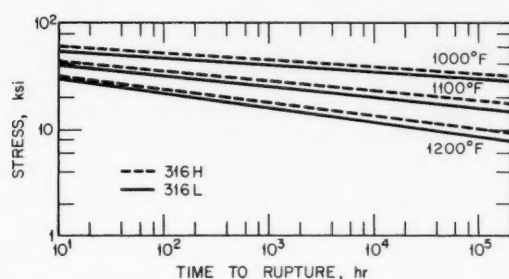
†From USAS B31.7.

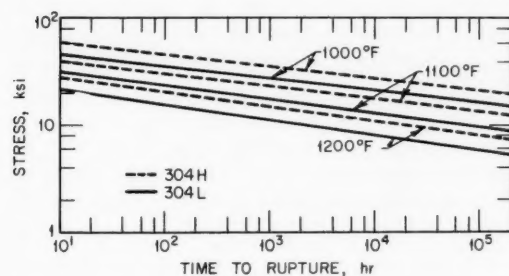Fig. 4 Design rupture strength for 316 stainless steel in air[10] (extrapolated from ASTM DS-5S2).

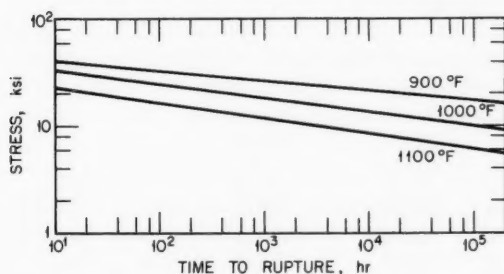Fig. 5 Design rupture strength for 304 stainless steel in air[10] (extrapolated from ASTM DS-5S2).

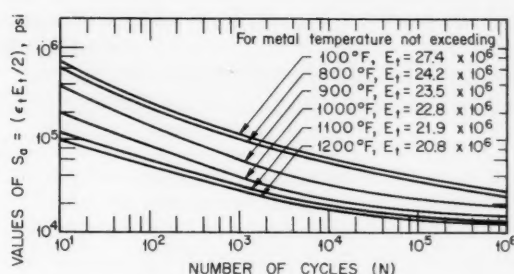Fig. 6 Design rupture strength for 2.25 Cr−1 Mo steel in air (extrapolated from ASTM DS 6S1).

Fig. 7 Design fatigue strength, S_a, for austenitic stainless steels in air. (ϵ_t and E_t are total strain range and Young's modulus, respectively.)
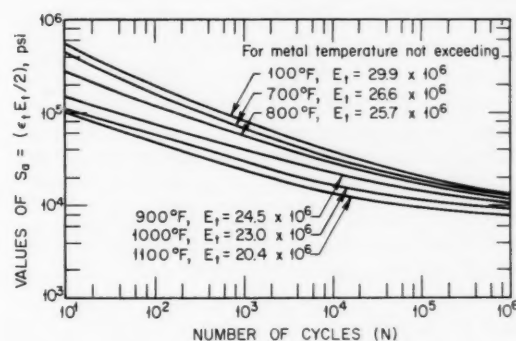

Fig. 8 Design fatigue strength, S_a, for 2.25 Cr−1 Mo steel in air. (ϵ_t and E_t are total strain range and Young's modulus, respectively.)

manual welding with coated electrodes. Rather, it must be made by the TIG process with a controlled inert-gas purge for inside surface protection.

All attachment welds used to join lugs, shoes, brackets, rings, or other nonpressure parts to the pipe wall, as well as welds joining branch connections or straight sections of pipe, should be full-penetration welds. The maximum depth of deposit in one pass should be ¼ in. for all types of welds except submerged-arc welds in material over 1 in. thick (which may have a ⅜-in. bead per pass). This limits the grain size in the welds.

Piping systems of austenitic stainless steel require no preheat or postweld heat-treatment. Parts made of 2.25 Cr−1 Mo steel require a 300°F (minimum) preheat before being thermally cut or welded and a postweld heat-treatment at 1300 to 1425°F for at least 2 hr.

Fabricators of LMFBR piping systems are responsible for conducting qualification tests for the welding procedures and for the performance of the welders in accordance with the stipulations in Section IX of the *ASME Boiler and Pressure Vessel Code*, with a few minor amendments as stipulated in the guide.

Examination, Inspection, and Testing

During construction each component of the piping system must be examined by a qualified representative

Table 10 Design Sustained Stress Intensity
for LMFBR Piping Materials

Temp., °F	Design stress intensity for sustained loading (S_{ms}), psi				
	304H	304L	316H	316L	2.25 Cr–1 Mo
Room	20,000	16,650	20,000	16,650	19,980
100	20,000	16,650	20,000	16,650	19,980
200	20,000	16,650	20,000	16,650	18,676
300	20,000	16,650	20,000	16,650	17,942
400	18,700	15,750	19,250	15,500	17,475
500	17,450	14,750	17,950	14,400	17,209
600	16,400	13,950	17,000	13,500	16,675
650	16,150	13,700	16,650	13,200	16,208
700	15,950	13,450	16,300	12,850	15,675
750	15,550	13,250	16,050	12,600	15,208
800	15,150	13,000	15,850	12,350	14,941
850	14,900	12,800*	15,700	12,100*	14,474
900	14,650	10,500*	15,550	11,800	14,074
950	14,350	8,500*	15,400	11,500*	9,500
1000	13,700	7,000*	15,300	11,200*	5,500
1050	10,800	5,600*	13,000	10,850*	4,100
1100	8,400	4,500*	10,400	7,400*	3,000
1150	6,720	3,500*	8,250	5,600*	
1200	5,280	2,800*	6,400	4,150*	

*Values given are for temperatures that exceed the limits of Section VIII of the ASME Code and USAS B31.7.

Table 11 Design Transient Stress Intensity
for LMFBR Piping Materials

Temp., °F	Design stress intensity for transient loading (S_{mt}), psi				
	304H	304L	316H	316L	2.25 Cr–1 Mo
Room	20,000	16,650	20,000	16,650	19,980
100	20,000	16,650	20,000	16,650	19,980
200	20,000	16,650	20,000	16,650	18,676
300	20,000	16,650	20,000	16,650	17,942
400	18,700	15,750	19,250	15,500	17,475
500	17,450	14,750	17,950	14,400	17,209
600	16,400	13,950	17,000	13,500	16,675
650	16,150	13,700	16,650	13,200	16,208
700	15,950	13,450	16,300	12,850	15,675
750	15,550	13,250	16,050	12,600	15,208
800	15,150	13,000	15,850	12,350	14,941
850	14,900	12,800*	15,700	12,100*	14,474
900	14,650	12,600*	15,550	11,800*	14,074
950	14,350	12,260*	15,400	11,500*	13,674
1000	14,040	11,920*	15,300	11,200*	13,073
1050	13,635	11,590*	15,120	10,850*	
1100	13,230	11,250*	14,850	10,575*	
1150	12,960	10,690*	14,715	10,000*	
1200	12,690	10,125*	14,580	9,450*	

*Values given are for temperatures that exceed the limits of Section VIII of the ASME Code and USAS B31.7.

of the owner for compliance with the design, as well as with the material, fabrication, assembly, installation, and test requirements. Where required by law, the piping-system components must also be examined by a qualified state, municipal, or insurance-company inspector. Nondestructive examinations must be conducted by duly qualified personnel using methods and acceptance standards given in Appendix B of USAS B31.7 for Class I piping.

Visual examinations of each weld are required before, during, and after welding, heat-treating, and testing to ensure that end preparation, dimensions, finish, alignment, fit-up, material, cleanliness, welding procedures, and weld quality are all in accordance with the specifications. All welds that contact sodium or contain pressure must be examined by 100% radiography. Completed weld surfaces, including the inside surface of the root pass of each accessible weld, should be examined for cracks or flaws by magnetic particle (for 2.25 Cr−1 Mo) or by liquid penetrant (for austenitic stainless steels). An exception is the outside of root passes in stainless-steel lines, which should not be given the dye-penetrant examination because any residual penetrant would cause contamination during the second pass.

Each section of pipe going into an LMFBR system must be hydrostatically tested by the manufacturer for at least 10 min at the design pressure or at 75% of the test pressure (whichever is greater), using high-purity water or other noncorrosive liquid. The many welds necessary to fabricate the pipespools* must be given either a hydrostatic test (as above) or a pneumatic pressure test by the fabricator and then leak checked with a helium mass spectrometer.

Finally the installed system is to be given a pneumatic pressure test at the design pressure or 75% of the test pressure (whichever is greater). For this test, the test pressure is defined as the lowest of the following:

1. The pressure that is 1.20 times the design pressure, multiplied by the lowest ratio of the allowable stress intensities at test temperature and at design temperature (from Table 10 or 11).

2. The pressure required to produce a calculated primary membrane stress intensity of 90% of the yield strength at test temperature (from Table 9).

3. The pressure required to cause the primary membrane plus primary bending stress intensity to

reach 135% of the yield strength at test temperature (from Table 9).

4. The allowable seat pressure on the main piping block valves or the relief capacity of the vents on the lower pressure portion of the system.

Hydrostatic testing of the installed system is not permitted because of the danger of leaving residual fluid that would be incompatible with the sodium in the system. Following the pressure test, the whole system must be leak checked with a helium mass spectrometer.

Cleanliness and Cleaning

An RDT standard (F 5-1T) entitled "Cleaning and Cleanliness Requirements for Nuclear Reactor Components" includes comprehensive requirements applicable to LMFBR systems. Stringent precautions are recommended during the fabrication, handling, storage, shipping, assembly, and testing of piping components and systems to prevent contamination that might have harmful consequences.

For example, antispatter and rust-preventive compounds should be carefully cleaned away from areas to be welded. After welding, flux, slag, and spatter should be removed, using sandblast on welds made with coated electrodes. Areas to be hot-formed or heat-treated should first be cleaned to remove all grease and paint, and a carburizing flame should be avoided. Only high-purity water (<25 ppM chlorides) should be used as the lubricant in fabrication and assembly operations involving piping components that contain crevices or areas that cannot be thoroughly cleaned later. Manometers or other equipment containing mercury or mercury compounds that could accidentally contact the piping should not be used. Clean components should be protected during fabrication and storage by the use of temporary plugs or seals (conforming to RDT F7-2T) to prevent contamination by such things as dust and salt air. Careful inventory control is needed to prevent tools, cloths, and small hardware from being left in the component. Hammers, pads, skids, and fixtures used during assembly and handling of stainless-steel piping components should not be made of such potentially harmful contaminating materials as Al, Zn, Cu, Pb, or babbitt metal.

Mechanical cleaning by grinding, polishing, brushing, filing, and deburring should be done with clean tools that have not been used on materials other than the piping material being cleaned. Only resin-bonded aluminum oxide grinding tools should be used on stainless steels, whereas rubber-bonded tools and sili-

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*A pipespool is a subassembly of pipe and fittings prepared in a shop to be field welded in place as a unit.

con carbide abrasives are also acceptable on 2.25 Cr–1 Mo steel. Brushing should be done with clean stainless-steel brushes, avoiding seal membranes or areas to be inspected with liquid penetrant. Stainless steel should be filed and deburred only with carbide tools. All particles resulting from the mechanical-cleaning operations should be carefully removed.

Water used in cleaning procedures must contain <25 ppM chlorides, have a conductivity of <400 μmhos/cm, a pH of 6.0 to 8.0, and no turbidity, oil, or sediment. No halogenated chemicals or lead-bearing or sulfur-bearing compounds should be used on stainless steel. Only clean acetone or alcohol is permitted for degreasing surfaces containing crevices. For surfaces without crevices or inaccessible areas, a non-ionic detergent (1 oz per gallon of water) or a hot (160 to 180°F) solution of trisodium phosphate (7 to 10 oz per gallon of water) and nonionic detergent (up to 1 oz per gallon of water), followed by drying, are also allowed. Drying should be done with clean, dry, oil-free air or inert gas, a drying oven, or evacuation.

Pipespool assemblies must be cleaned in accordance with the RDT F 5-1T specifications for their particular class. Since an LMFBR system will be complex and have a low tolerance for residues of chemical cleaning solutions, the cleaning of an installed system poses extremely difficult problems. To minimize these problems, the pipespools must be packaged, shipped, stored, and assembled in a manner that will maintain their cleanliness.

To date the most practical method found for producing a clean, finished sodium system is to assemble it from carefully cleaned components, with the least contamination practicable during installation, and then to circulate hot sodium through it to getter whatever contaminants are left. If proper care is exercised in cleaning and installing the pipespools, the filters and impurity traps in the sodium loops should have no difficulty reducing the sodium impurities to acceptable levels following the initial system cleanup. This has been borne out in a number of sodium facilities that have been built and operated previously.

HEATING AND INSULATION

Heating Methods

Sodium in tank cars or storage tanks will be heated to at least 350°F to prevent freeze-up during charging into the LMFBR system. A sudden flow of 350°F sodium into an empty pipe at room temperature would produce significant thermal stresses in the pipe wall as a consequence of the resulting large thermal gradients. To alleviate this condition, some means of preheating the pipe must be employed to bring its temperature close to that of the sodium when flow is initiated.

Because the sodium circuits built to date have had widely varying heating requirements, each one was equipped with a different custom-designed pipe-preheating system. For the same reason the guide makes no attempt to standardize LMFBR preheaters. However, since they have some effect on the piping design, it is important that the piping designer knows the characteristics of the various systems available and selects the one that best suits his needs.

On the basis of a comparative study,[12] C. F. Braun and Company described the following types of pipe heaters and rated them as shown in Table 12.

1. *Induction heaters* induce eddy currents in a pipe of magnetic material by an alternating current passing through a coil wrapped around the pipe. For (nonmagnetic) austenitic stainless-steel pipe, a carbon-steel jacket is placed between the coil and the pipe. Generally the coil is placed outside the insulation. Efficiency is poor.

2. *Transformer and direct-series heaters* both force current to flow through the pipe itself, thus generating heat from the resistance of the pipe itself. In direct-series heating, electrodes are attached to the pipe, causing compatibility, insulating, and thermal-stress problems. In transformer heating the pipe runs through a transformer core and becomes the secondary winding with no direct electrical connections.

3. *Tubular resistance heaters* consist of helically coiled wire centered in a metal sheath with compacted mineral insulation between. These elements are usually run parallel to the pipe, with thermal insulation covering both pipe and heater. Problems may stem from hot spots due to coil bunching at pipe bends, mechanical damage to terminals, or inability to dissipate enough generated heat to prevent burnout.

4. *Mineral-insulated sheathed-cable resistance heaters* are constructed like tubular heaters, but with a noncoiled heating element. They come in much longer lengths (500 ft vs. 50 ft) and have about the same drawbacks, except bending is not as serious a problem.

5. *Cartridge-type resistance heaters* are short tubular heaters (12 in. maximum) which have both terminals at the same end, have higher power densities, and are used to heat a localized area.

6. *Strip-type resistance heaters* are like flattened tubular heaters containing one or more heating elements up to 10 ft long. They are designed for flat surfaces or large-diameter pipes.

Table 12 Preheater Ratings* for LMFBR Piping[1][2]

Heater type	Evenness of preheat to 350–400°F (or full pipe to 800°F)	Reliability at pipe temperatures up to 1200°F(3)	Control-lability	Materials compati-bility(5)	Ease of inspection	Resis-tance to mechanical damage	Ease of assembly and dis-assembly	Extent of contribution to stress
			Electric Heaters					
Induction	F(1)	F	F	G	P	P	P	G
Transformer	P	P	P	F	G	G	G	G
Direct series	P	P	P	F	G	G	G	(4)
Tubular	(4)	G(6)	G(4)	G	G(4)	G(4)	F(4)	(4)
Cable	(4)	G(6)	G(4)	G	G(4)	G(4)	F(4)	(4)
Cartridge	P	G(6)	G(4)	G	G(4)	G	G	(4)
Strip	(4)	P	G(4)	G	G(4)	G(4)	F(4)	(4)
Tape	(4)	G	G(4)	G	G(4)	G(4)	F	(4)
Mantle	G(4)	G	G(4)	G	P(4)	G(4)	F	G
Blanket	G(4)	G	G(4)	G	P(4)	G(4)	F	G
Radiant	G	G	G(4)	G	F	F	F	G
			Fluid Heaters					
Annular	G(2)	G	F(4)	G	F	G(4)	G	G
Trace	(2)(4)	G	F(4)	G	G	G(4)	F	(4)
Furnace cell	G	G	P	G	G	G	G	G

*G = good; F = fair; P = poor; ratings are based on typical designs.
(1) May require jacket.
(2) Limited by fluid temperature.
(3) Dependent on materials.

(4) Dependent on installation.
(5) Ratings based on available materials.
(6) Qualitative information indicates method is good when properly installed; quantitative data are lacking.

7. *Flexible resistance tapes, mantles, or blankets* are composed of small-gauge Nichrome wires insulated with $SiO_2-Al_2O_3-MgO$ or metal-sheathed MgO and woven with Fiberglas, silica compound, or stainless-steel mesh into tapes. Mantles or complete blankets are made by sewing the tapes into Fiberglas insulation.

8. *Radiant electric heaters* generally consist of high-temperature resistance heaters installed a short distance from the surface to be heated and thermally insulated and shielded so as to radiate toward it.

9. *Annular fluid heaters* consist simply of an annular jacket around the sodium lines and a blower for forcing hot air or other hot gas through the jacket. Manifolding, distribution, and control are difficult for complex piping systems.

10. *Fluid trace heaters* are similar to annular heaters, except that the hot fluid, such as oil or air, is carried through a narrow pipe next to the sodium line instead of through an annulus.

11. *A furnace-cell heater* is an insulated enclosure completely engulfing the piping system, with a heated atmosphere inside. Heating is uniform and no insulation is needed on the piping, but local control is impossible.

Insulation

As with insulation for conventional piping, LMFBR pipe-insulation materials must have a low thermal conductivity, ability to withstand high temperatures, and be unreactive with the piping and the piped fluid. It is particularly important that they be nonhygroscopic because of the extreme reactivity of sodium with water. The presence of preheaters between the insulation and the pipe wall means that the insulation may have to withstand concentrated temperatures that are higher than that of the piping.

Frequently a stainless-steel sheet is installed to serve as both a physical and thermal baffle between the preheater and the insulation. In the event of a sodium leak, this sheet could retard permeation of the insulation by the sodium.

Installation and replacement could be facilitated if the insulation were formed into prefabricated blocks or encased in preformed sheet metal, perhaps even with integral preheaters incorporated. The insulation should be held in place by external straps. Studs or support rings welded to the pipe wall are not permitted.

Materials that withstand high service temperatures frequently have higher thermal conductivities than the less heat-resistant insulations. For this reason, two different materials are sometimes used in layers. The following materials are considered suitable as pipe insulation for most LMFBR applications:[12]

Material	Max. service temp., °F	Density, lb/ft³
Unbonded Fiberglas	1000	9–11
Calcium silicate	1200	11
Amosite asbestos	1500	16
Reflective insulation	1500	
Diatomaceous silica	1900	24
Mineral wool	1900	17
Ceramic wools	2300	3–24

IN-SERVICE SURVEILLANCE

A study of piping-failure experience[13] revealed that many failures could have been avoided or detected before becoming catastrophic if adequate surveillance methods had been used. A piping failure during full-power operation of the LMFBR would have such serious consequences that elaborate, reliable in-service surveillance provisions are not only justified but also essential for safety.

Devices must be incorporated to monitor for sodium leaks, cracks in the piping components, adverse environmental effects on the materials, and pipe movements during and after the initial sodium fill. The recommended techniques were selected following a state-of-the-art survey[14] by C. F. Braun and Company.

Sodium Leak Detection

The primary piping in an LMFBR will be either single or double walled, contain highly radioactive sodium, and be located in galleries and surrounded by an oxygen-depleted or inert-gas atmosphere that is either circulated by blowers or naturally convected through coolers to keep the temperature of the piping at a tolerable level. The secondary piping is expected to be single walled only, contained in galleries filled with oxygen-depleted air in some zones, and exposed to atmospheric air in other zones. Any sodium leak-detecting system must be selected specifically for the type and configuration of the piping it is intended for, and backup detectors should be installed whenever feasible.

Radiation-Type Detector. Since the sodium in the primary system will become radioactive, a device that responds to this activity could be used to detect a leak. Commercial instruments of this type are available, one of which employs a fan to force the air (or inert gas) from the galleries surrounding the piping through a strip of filter paper that is advanced continuously and monitored for radioactivity. Any beta or gamma radiation from condensed sodium vapor or sodium oxide particles collected on the filter activates an alarm. If gallery cooling of single-containment primary piping is by forced circulation, only one such detector is needed for the entire primary system. With natural-convection cooling, several detection units are necessary.

Because of its high sensitivity, a radiation detector should be effective in spite of the large dilution of the escaped radioactive sodium vapor or oxide particles by the circulating gas in the galleries. Leaking radioactive sodium will be hindered by the pipe insulation and heaters from mixing with the gas stream. Sensitivity can be improved by bleeding some of the gas through purposely installed small passages between the insulation and the pipe.

Radiation-type leak detectors are, of course, useless for detecting nonradioactive sodium leaks. This means that they cannot function during initial nonnuclear shakedown of the plant, nor can they be used in secondary loops or service systems. Also, it would be awkward to use them in plants that have double-containment primary piping since the inert gas in the annulus is not normally circulated. Radiation-type leak detectors also restrict the gas temperature to about 120°F and limit the sensitivity, depending on the design. The commercial monitor described quotes the minimum detectable activity as 7×10^{-12} Ci/m³ with an air flow of 10 cfm and a filter tape speed of 1 in./hr.

Resistivity-Type Detector. There are several detector designs that depend on measuring changes in resistance resulting from the presence of sodium or sodium compounds. About the simplest, most rugged, and inexpensive detector consists of a pair of wires which are insulated electrically from each other but are so located that sodium leaking out of a pipe or fitting would short them together. One such design is based on wires strung through special two-hole ceramic beads with cutouts to permit the sodium to reach the wires with ease. Another design is in the form of a tape made up of two wires with porous, woven high-temperature insulation that is easily penetrated by leaking sodium.

Such detectors are heat resistant and can be placed directly against the pipe or over the preheaters. These detectors, although they are among the best developed

to date for single-containment primary and secondary piping, have several shortcomings. At temperatures below about 600°F, sodium may not wet the conductors or penetrate the electrical insulation; because of the ease with which the wires can be shorted unintentionally, false alarms are likely; the pipe orientation, shape, and thermal insulation may prevent leaking sodium from finding its way to the detector; an excessive number of sensors would be required to achieve a high probability of detecting a small leak promptly; small leaks may not produce any response because of excessive oxide formation and confinement of the sodium; accessibility for repair in the event of malfunction is rather poor.

A different kind of resistivity-type leak detector is recommended for double-containment primary piping. A gas sample drawn from the annulus between the inner and outer pipes is bubbled through water. Any sodium vapor or sodium oxide in the gas will go into solution and decrease the electrical resistivity of the water. This detector is simple, inexpensive, and easy to service since the only equipment needed is a sampling tube (or several), a bubbler, and a device for tripping an alarm when the resistivity of the water in the bubbler drops below a preset value. A disadvantage is that the detector may have a long response time for a small leak since it depends on diffusion of the leaking sodium throughout the annular regions. Also, the detector removes gas from the annulus, necessitating makeup and increasing the probability of contaminating the inert gas.

Level-Type Detector. A device considered suitable as a backup detector to the aqueous resistivity type for double-containment primary piping consists simply of a spark plug installed in the lowermost sections of the outer piping where sodium can collect in the event of a leak and short the center electrode to the case, tripping an alarm. Response time varies with the proximity and magnitude of the leak. A disadvantage of the spark-plug device is that its installation creates a potential weak spot in the double containment. A leaking spark plug would permit air contamination of the inert gas in the annulus.

The spark-plug-type detector is also a backup candidate to the two-wire resistance type for the secondary system. For this application, probe-equipped collectors would have to be installed against the insulation but below the piping. Channels through the insulation could be provided at appropriate locations to keep the insulation from blocking the flow of leaking sodium. With small leaks this detector would

either have a very long response time or not work at all.

Optical-Type Detector. Since this device depends on the attenuation of a light beam by sodium vapor or smoke, it is considered suitable for detecting leaks in secondary-system piping in air. A suggested way to facilitate the egress of smoke resulting from the sodium–air reaction in the event of a leak is to provide channels between the pipe and insulation or through the insulation. Suction can be applied at the channel exits to evacuate the smoke and exhaust it through a chamber containing the smoke detector. Limiting bulk sensitivities to sodium vapor and smoke are 75×10^{-6} and 1.5×10^{-3} g/m^3, respectively. The main drawback of this device is that it has high sensitivity and rapid response only for leaks in the vicinity of the detecting beam. In a large complex system, many instruments would be needed to obtain satisfactory coverage.

Crack Detection

Once an LMFBR goes into shakedown testing, it is highly desirable to be able to detect and follow the growth of a defect in any piping component from its onset or initial (acceptable) size toward the stage at which it becomes dangerous. Techniques for determining the growth rate of a crack and for predicting the initiation of failure with reasonable accuracy would be of great value in eliminating catastrophic accidents once the plant has begun operating.

The three general methods of crack or flaw detection considered most promising for this purpose are optical, ultrasonic, and acoustic emission. Unfortunately, at their present state of development, all three methods require substantial additional development and adaptation before they can be applied routinely to LMFBR piping.

Optical Methods. The radioactive environment of the primary system excludes it entirely from direct visual observation in service; whereas the secondary system, hidden by insulation and operating at high temperatures, would be most difficult to observe directly. Closed-circuit television monitors would solve some of the direct-observation problems, but it would be economically prohibitive to locate television cameras at every potential trouble spot. Picture resolution would have to be outstanding to be useful. The cameras could not be used under sodium, and even in air their temperature would have to be kept below 150°F.

Optical borescopes and periscopes can be installed to scan areas in remote and inaccessible places. The

borescopes and periscopes give sharp images of properly prepared complex surfaces, and they are not unduly sensitive to temperature. Although these devices cannot be used under sodium, they offer at least limited potential for scanning the outer surfaces of LMFBR piping components.

Ultrasonic Methods. In the ultrasonic pulse–echo method, a pulse is generated in the pipe material, and echoes from internal flaws are picked up by a receiver. Although highly successful in the shop, this method poses a number of difficulties for in-service surveillance of LMFBR piping: (1) the transducer has to scan the surface and yet must be located with great precision; (2) a method of producing continuous records for comparison with earlier scans must be developed; (3) much skill and experience are needed to operate the equipment and interpret the results.

Acoustic-Emission Methods. When a flaw or crack in a pipe propagates, energy is released in the form of elastic waves that can be detected with transducers fastened to the pipe walls. The outputs of a number of strategically located transducers can be triangulated to locate the crack and indicate its size and growth rate. Although acoustic-emission transducers are not yet perfected for LMFBR conditions, the high-temperature piezoelectric type with a lithium niobate sensor appears to be quite promising.

PIPING-MATERIALS SURVEILLANCE

In-service surveillance of the previously discussed effects of the LMFBR coolant-system environment on the piping materials is considered necessary to (1) ensure that the material strength properties have not been dangerously degraded and (2) provide information for predicting the remaining service lifetime of the piping. The favored technique involves placement of specimens of the piping materials into flowing sodium environments under conditions duplicating those to which the piping itself is subjected at selected exposure sites. Periodically, selected specimens are removed and tested to obtain corrosion and strength data for comparison with preexposure properties. Obviously in-service materials surveillance for the reactor vessel, internals, and major components will have much in common with piping-materials surveillance, and close coordination of plans and facilities will be required.

As indicated earlier, impurities in sodium can have large effects on corrosion and mass transfer and must therefore be monitored continuously in conjunction

with any in-service materials-surveillance program. Purity indicators are generally located in small-diameter branch lines. Some of the instruments used or being developed for this purpose are hydrogen meters, carbon meters, and three types of oxygen meters— plugging meters, rhometers, and electrochemical oxygen meters.

Specimen-Exposure Systems. Two types of specimen-exposure systems are being considered for materials surveillance: the direct-insertion system and the bypass system. Either system should permit groups of test specimens to be inserted and removed without contaminating the sodium or the specimens, without hazard to personnel, and without interfering with continuous full-load plant operation. The design should permit each group of specimens to be inserted or removed independently of the others, allow the radioactive sodium to drain from the specimens prior to removal, and provide for instrumentation to monitor sodium temperature and flow rate. The design, however, should not introduce any new materials into the piping system.

To insert specimens directly into the main primary and secondary sodium pipes requires that specimen insertion and removal facilities be welded to the piping at selected sites in the hot leg and cold leg of each loop. A major advantage of the direct-insertion system is that it exposes the test specimen to very nearly the same coolant environment (Reynolds number, downstream conditions,* and sodium chemistry) as the piping at the exposure sites. The main disadvantage of this system is its complexity and the fact that it jeopardizes the piping-system integrity.

In the bypass method of specimen exposure, test specimens are inserted in fixtures in auxiliary bypass loops tapped into the main sodium piping at sites representing various environmental conditions. Freeze seals and isolation valves in the bypass piping permit the specimens to be inserted or withdrawn with minimum risk to the integrity of the main piping. The disadvantage of this system is that the environment to which the specimens are exposed is not identical to the environment at the main-piping site. This is especially likely to be true of the Reynolds number and downstream conditions.

Test Specimens. Insofar as possible the test specimens should be made from the same heats of materials

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*"Downstream conditions" refers to the fact that for a given sodium temperature and flow velocity, the corrosion rate diminishes in the downstream direction.

and subjected to the same forming, heat-treatment, fabrication processes (including welding), and cleaning processes as used for the actual LMFBR piping. Each weld-type specimen should include some unaffected base metal along with the heat-affected zone.

Specimens should be of forms suitable for tensile, stress-rupture, creep, fatigue, and impact testing, as well as for chemical, microstructural, and weight-loss evaluations. Because of the large number needed, subsize specimens conforming to available ASTM, RDT, and other applicable standards should be used whenever possible. For example, assuming 10 specimens each are required to characterize a base metal, a similar-metal weld, and a dissimilar-metal weld, a system with a bimetallic cold leg would require at least 420 cold-leg specimens and 210 hot-leg specimens to accommodate evaluations at 1, 2, 5, 10, 15, 20, and 25 years after plant startup.

Sufficient counterpart specimens should be provided for preexposure testing and examination, for comparison with postexposure results. ASTM recommendations should be used for tensile (E8 and E184), stress-rupture (E139), creep (E139), and impact (E23 and E184) tests.

Monitoring of Pipe Movements

The design analysis will have established predictions of piping-system deflections resulting from anticipated thermal-expansion and loading variations during LMFBR operation. It is recognized that the system may behave quite differently for a variety of reasons (local yielding, nonisothermal behavior, variations in thermophysical properties of the materials, nonuniformity of wall thickness, out of roundness, simplifying assumptions in the flexibility analysis, etc.).

To evaluate the effects of the actual deflections, including the strain cycling (fatigue) from day-to-day variations in temperature and load, in-service surveillance of pipe movements is most desirable. Such monitoring should be capable of detecting interference between piping components and structures, improper functioning of pipe supports, and excessive vibrations, as well as predicted and unpredicted thermal strains. Various techniques, both optical and mechanical, are being considered for measuring pipe movements while the system is in operation.

In accessible, nonradioactive areas, direct displacement-time measurements can be made visually, using suitable bench marks. In less accessible places, closed-circuit television can be used, provided the temperature is below 150°F and the tube can be replaced when needed. Borescope and periscope observations are suitable for otherwise inaccessible piping. Mechanical linkage systems, used in conjunction with remote displacement transducers such as dial indicators or differential transformers, can detect relative displacements with extreme accuracy, provided temperature changes are either compensated for or eliminated.

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Sodium-Cooled Reactors: Rapsodie Physics Aspects

By Myrna L. Steele*

Rapsodie [the name is derived from the French words for fast (rapide) and for sodium] is a two-loop sodium-cooled fast reactor located at the Cadarache Center for Nuclear Studies in southeastern France about 40 miles north of Marseilles.

The original core loading was designed for a 20-MW(t) power level, which was reached in May 1967. The power level was increased to 24 MW(t) in December 1967. Early in 1970 the original core was replaced by a modified version, and on Aug. 13, 1970, a power level of 41 MW(t) was reached. As the reports on the physics measurements with this modified (Fortissimo) version become available, they will be summarized in an attempt to keep the information on Rapsodie as current as possible.

The first core[1] loading (shown in Fig. 1) was comprised of hexagonal subassemblies that contained either mixed-oxide fuel rods or depleted-uranium-blanket rods. The central core region consisted of 64 fuel subassemblies with each subassembly containing 37 fissile (mixed UO_2 –PuO_2) pins and 7 depleted UO_2 pins; the 500 blanket subassemblies, which surrounded the central core region, each contained only 7 pins of depleted UO_2. The core—blanket region was surrounded by two rows of stainless-steel-reflector subassemblies; the six B_4C control rods were located in the fourth row.

Loading of the original Rapsodie core was begun on Jan. 20, 1967, and criticality was attained on January 28 with 52 fuel subassemblies. Low-power testing was started on February 4, after a brief shutdown for

resetting some operations instruments, and continued through August 9. The central core region had a maximum neutron flux[5] of 1.7×10^{15} neutrons/$(cm^2)(sec)$ and sodium inlet and outlet temperatures of 410°C and 500°C, respectively.

The results of the physics tests on the initial core are discussed in this article. For discussion purposes the tests are divided into two groups: (1) basic physics (neutronics) characteristics and (2) integral physics parameters that relate to reactor operation. The first group consists of neutron fission- and capture-rate distributions, neutron fluxes and spectra, and gamma heating rates; the second group includes isothermal temperature coefficients of reactivity, power coefficients of reactivity, and rod calibrations.

NEUTRONICS CHARACTERISTICS

Fission- and Capture-Rate Measurements

During the low-power testing of Rapsodie, fission foils[2] were used for mapping the fission rates along the core and blanket subassemblies. Count data were obtained from the fission foils by the familiar two-energy-band technique.† The axial and radial distributions of the fission rates are shown in Figs. 2 and 3 for ^{235}U, in Figs. 4 and 5 for ^{238}U, and in Figs. 6 and 7 for ^{239}Pu; the axial capture-rate distributions for ^{238}U are shown in Figs. 8 and 9.

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†For a brief explanation of this technique, see Sodium-Cooled Reactors: Physics Aspects of the Enrico Fermi Fast Breeder Reactor, *Reactor Technology*, 13(3): 191-213 (Summer 1970).
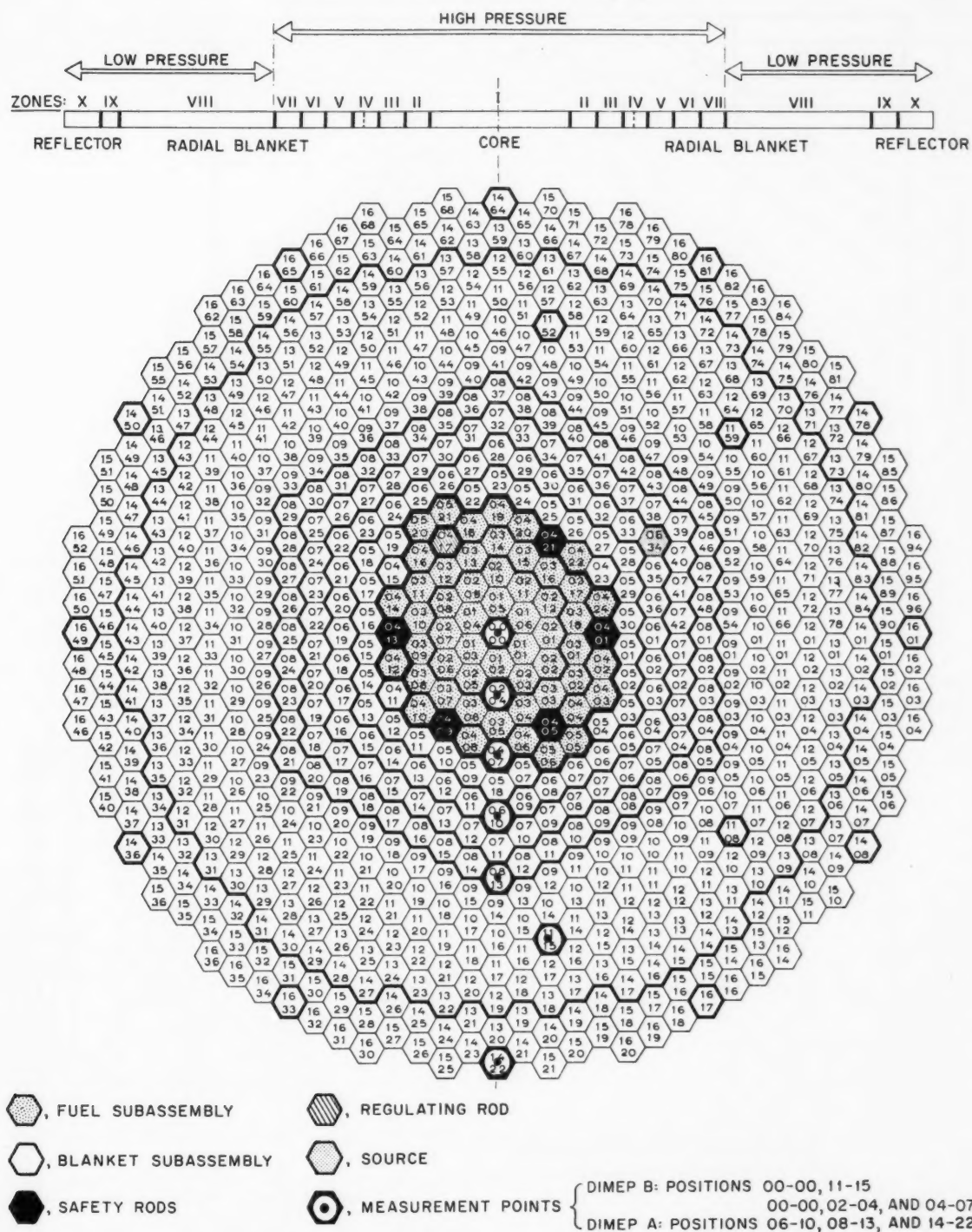
Fig. 1 Loading diagram and measurement points for startup testing.[1]
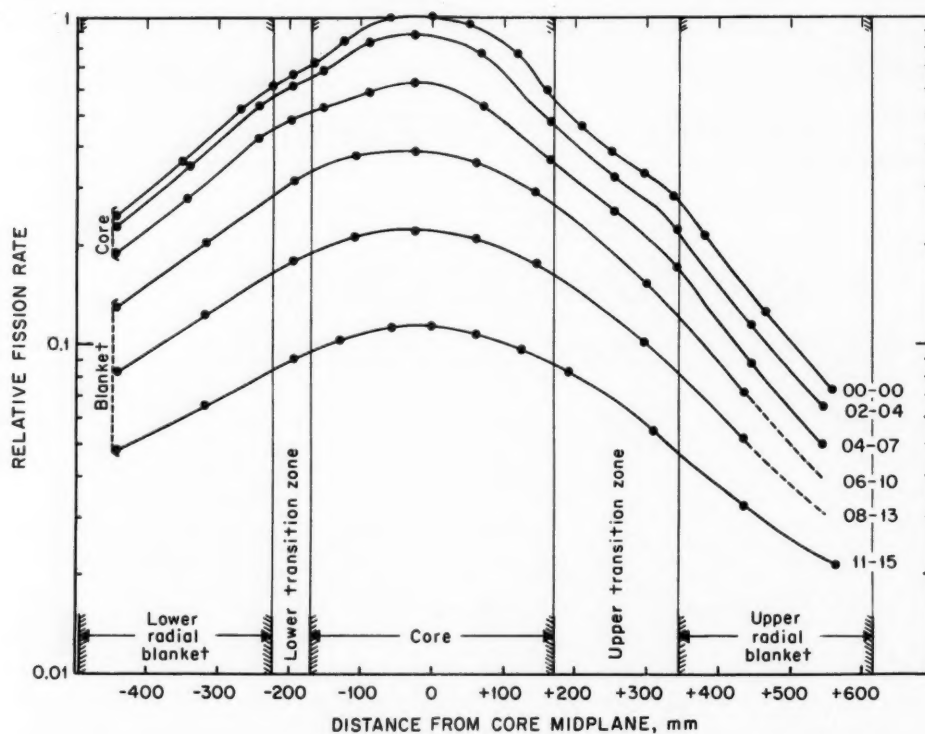
Fig. 2   Axial fission-rate distributions[2] for ^{235}U
(in positions 00–00, 02–04, 04–07, 06–10,
08–13, and 11–15).
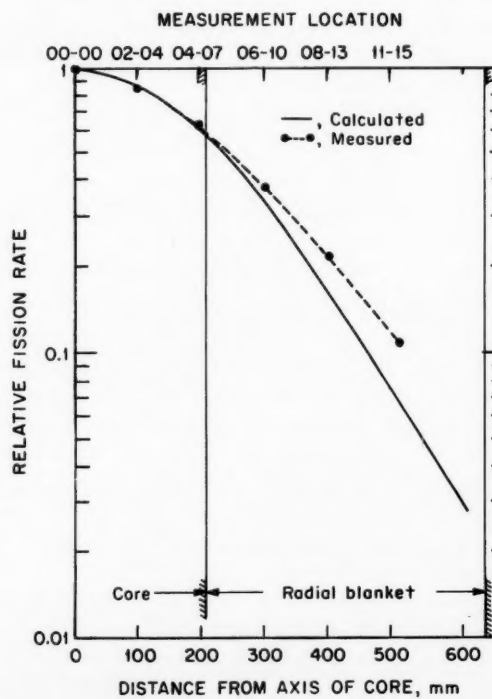
Fig. 3   Radial fission-rate distributions[2] for ^{235}U.
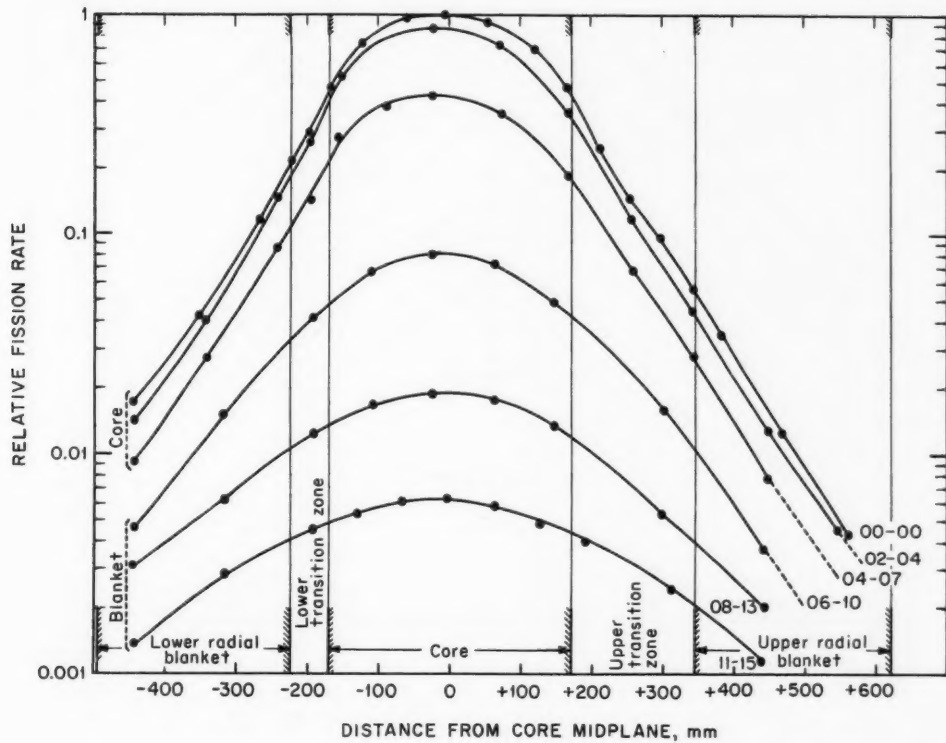
Fig. 4   Axial fission-rate distributions[2] for ^{238}U (in positions 00–00, 02–04, 04–07, 06–10, 08–13, and 11–15).
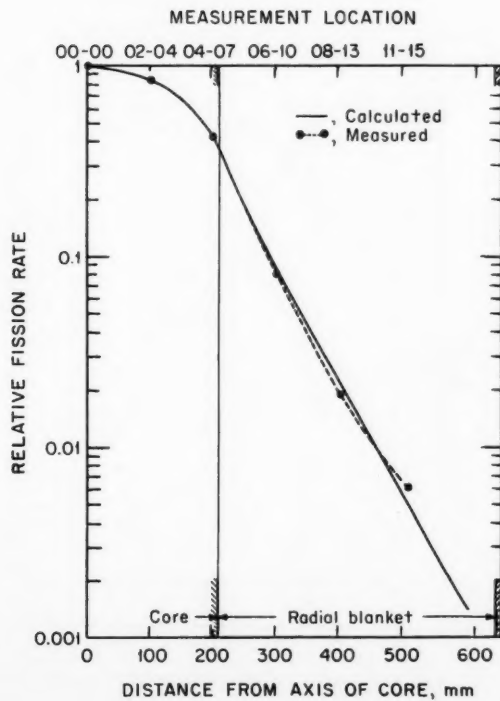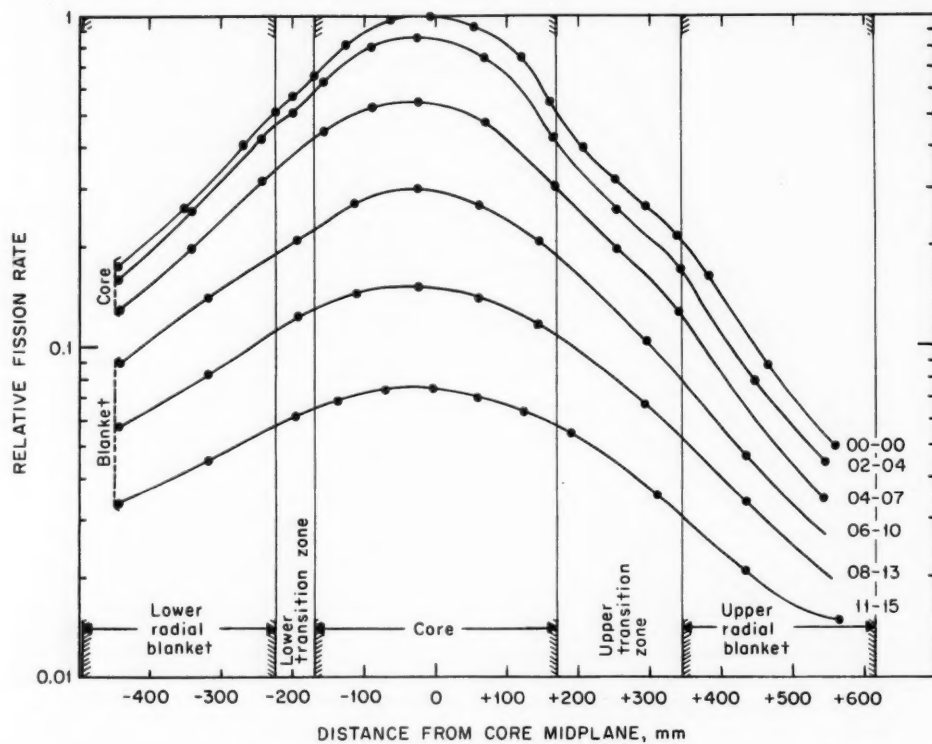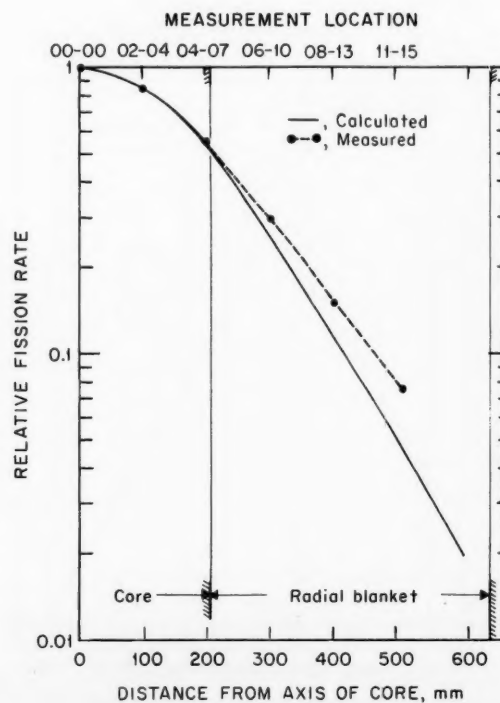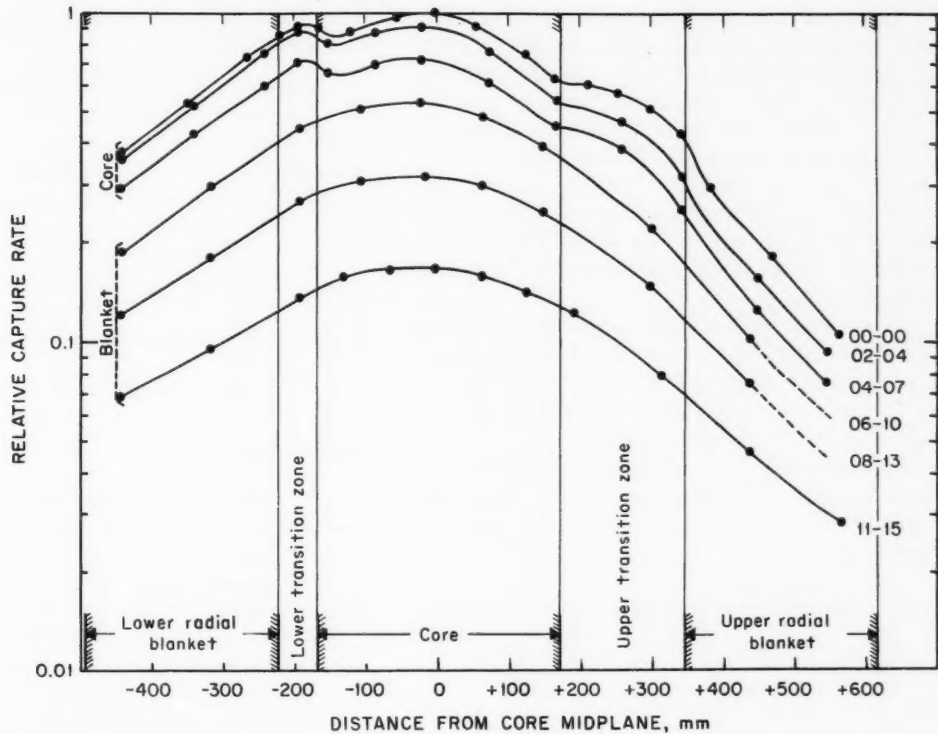

Fig. 5   Radial fission-rate distributions[2] for ^{238}U.

Fig. 6 Axial fission-rate distributions[2] for ^{239}Pu (in positions 00–00, 02–04, 04–07, 06–10, 08–13, and 11–15).

Fig. 7 Radial fission-rate distributions[2] for ^{239}Pu.

Fig. 8 Axial capture-rate distributions[2] for ^{238}U (in positions 00–00, 02–04, 04–07, 06–10, 08–13, and 11–15).
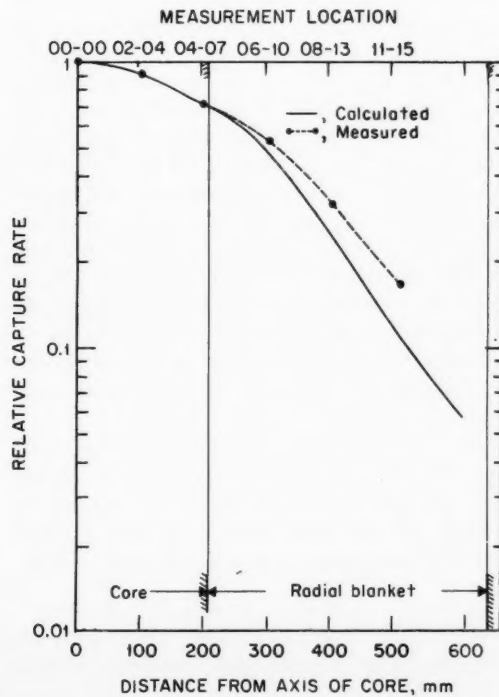
Fig. 9 Radial capture-rate distributions[2] for ^{238}U.

Additional fission-rate data were obtained with foil-type activation detectors of Mn, Au, Rh, and Ni. The foils were irradiated in special test subassemblies (DIMEPs) that were designed and fabricated to simulate core, blanket, and reflector elements. Figure 10 shows the axial fission-rate distributions that were obtained with the Mn, Au, ^{238}U, Rh, and Ni; the radial fission-rate distributions are shown in Fig. 11 for these detectors. The data in Figs. 10 and 11 have been normalized to 1 at the core center to facilitate comparison.

Fission chambers were also used to measure fission rates at the center of the core. The fission chambers had active areas composed of ^{235}U, ^{239}Pu, ^{237}Np, or ^{238}U. These measurements are compared in Table 1 with those obtained using fission foils.

Spectral indices were also obtained from the fission-foil data. Calculated spectral-index values are compared with results of fission-foil measurements in Table 2.

Neutron-Flux Maps of Core, Blanket, and Reflector

Detailed flux maps[3] of the core, blanket, and reflector were made with the Mn, Au, Rh, S, and Ni activation foils that were irradiated in the special DIMEP test rigs. All measurements, with the exception of the nickel, were made on a 56-subassembly core immediately after initial criticality. Measurements using nickel foils were made later in the reflector of a 61-subassembly core. Table 3 compares the calculated flux values obtained from a 52-energy-group NIOBE code with the average measured flux values at the core center at a reactor power level of 20 MW(t).

Gamma Heating Measurements

Calorimeters,[4] composed of five stainless-steel or graphite samples with an electric heater and thermocouple attached, were used to measure the gamma heating rates at several power levels along the central axis of the core and in an experiment channel in the 11th row. Results of these measurements are given in Table 4.

INTEGRAL PHYSICS PARAMETERS

Reactivity Measurements

Isothermal Coefficient. The isothermal temperature coefficient[5] of reactivity was measured between 160 and 430°C at the start of the Rapsodie testing program. Results of the measurements (Fig. 12) show

that the average value of the isothermal temperature coefficient was -3.95 ± 0.035 pcm*/°C.

Power Coefficient. Power coefficients were measured in March 1967 during the first approach to power and in June 1967 during the physics testing program. The combined results (Fig. 13) show that at nominal flow the power coefficients were

$$-24.6 \pm 1.7 \text{ pcm/MW at 0 to 5 MW}$$

$$-31.6 \pm 1.7 \text{ pcm/MW at 10 to 20 MW}$$

and at one-half nominal flow the power coefficient was

$$-41 \pm 2 \text{ pcm/MW at 0 to 10 MW}$$

Control-Rod Calibrations

The original Rapsodie core was equipped with five safety—shim rods and one regulating rod positioned symmetrically around the active zone of the core;[5] however, provisions had been made for two regulating rods, one heavy and one light. The poison portion of the rods was 450 mm long, and the absorber section of the rods contained B_4C that was 90% ^{10}B. The control rods in the first core loading were calibrated by positive-period, subcritical count-rate, and rod-drop measurements.

Positive-period data were obtained by varying the reactivity and recording the subsequent changes in power level on a 400-channel recorder. The changes in reactor power were recorded over 400-sec intervals for various reactor periods between 50 and 200 sec; these results were then plotted and analyzed graphically. Figure 14 shows the reactivity worths as a function of rod height both for the heavy regulating rod (which contains 21% ^{10}B) and for the light rod (which contains 11.5% ^{10}B). The total worths of the rods were 179 ± 6 pcm for the standard rod and 280 ± 6 pcm for the heavy rod. In Fig. 15 the reactivity worths of safety rods 1 and 4 are compared as a function of rod height; the individual worths of each whole rod were measured as 1641 ± 40 and 1603 ± 40 pcm, respectively.

Rod-drop tests were also conducted to assess reactor behavior subsequent to (1) dropping one rod (safety rod No. 1) from the full-out position and (2) dropping the rod bank from a partially inserted position that corresponded to a 20 MW(t) power level.

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*The term pcm means pour centimille, a unit used by the French to express reactivity. For conversion purposes, 1 pcm = 1×10^{-5} $\Delta k/k$.
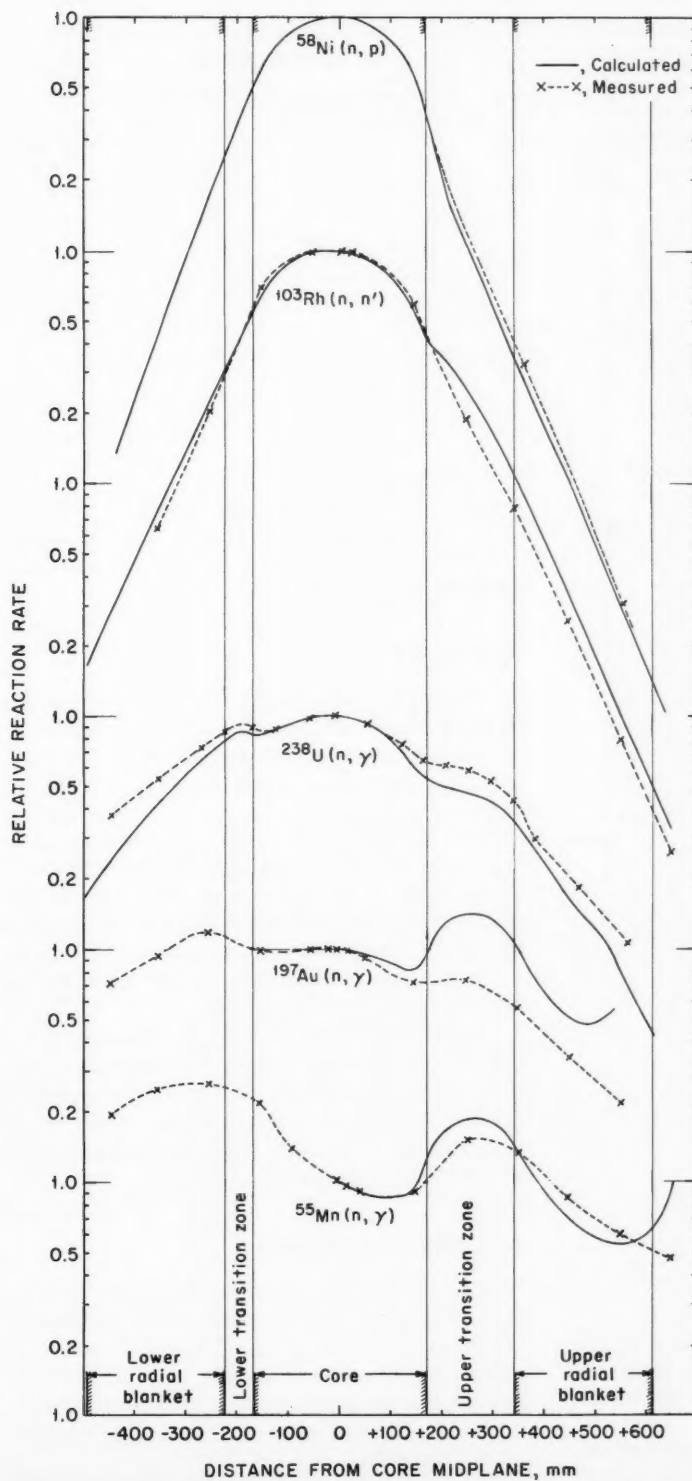
Fig. 10 Axial fission-rate distributions[2]
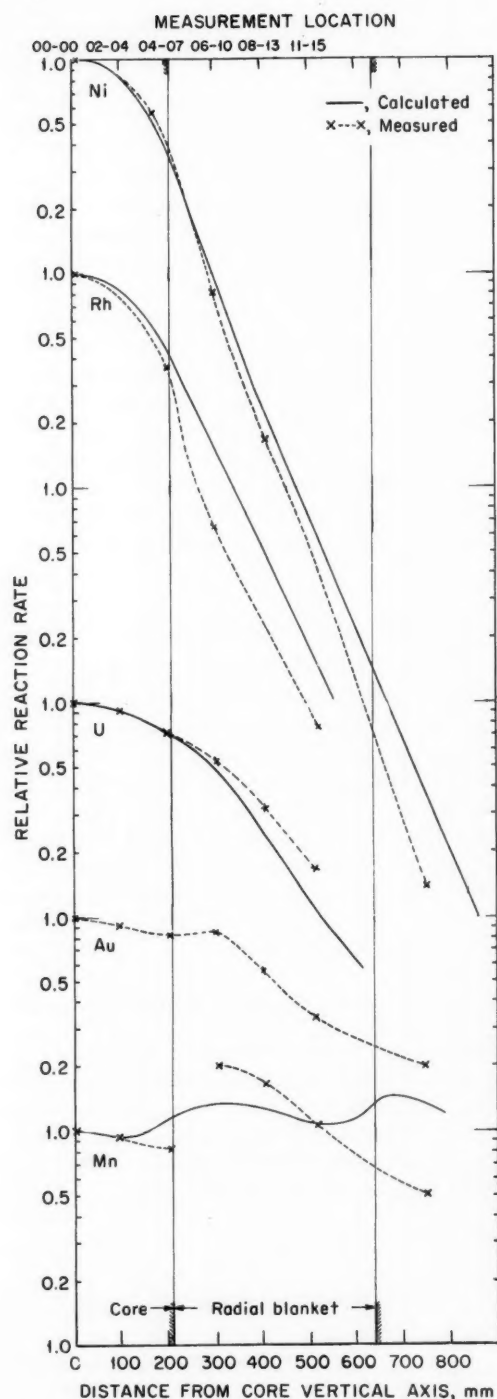for activation detectors.

Fig. 11 Radial fission-rate distributions[2] for activation detectors.

Table 1   Absolute Fission and Capture Rates at Core Center[2]

(56 Fuel Subassemblies; Temperature at 160°C)

	Fission chamber	Fission foil
Fission rate, 10^3 fissions/(g)(sec)(W)		
^{235}U	322	342
^{239}Pu	407	430
^{238}U	25.0	25.1
Capture rate, 10^3 captures/(g)(sec)(W)		
^{238}U		36.3

Results of the rod-drop tests combined with the safety-rod calibration curves (Fig. 15) gave the level as a function of time (Fig. 16). Results of the rod-drop measurements are summarized in Table 5.

SUMMARY

This is the third in a series of articles on the physics characteristics of the presently operating liquid-metal-cooled fast breeder reactors (LMFBRs). In the $2\frac{1}{2}$ years that Rapsodie has been operating, it has significantly contributed to the world's knowledge of LMFBR technology. These very significant contributions have led me to undertake the translation and condensation of the reports used as the basis for this article. I hope to keep the material on Rapsodie as current as possible in future issues so that cross-comparisons of LMFBR data can be made by the reader on a relatively current basis.

Readers who need more detailed information are urged to get the reports listed in the Reference section or to write to Rapsodie personnel at the Commissariat a l'Énergie Atomique, Centre d'Études Nucléaires de Cadarache, Départment de Recherche Physique, Boite Postale N° 2, Gif-sur-Yvette, 91 France.

ACKNOWLEDGMENTS

Table 2   Calculated and Measured Spectral Indices[2] for Rapsodie

Spectral index	Measured		Calculated				
	Fission chamber*	Detector	(1)	(2)	(3)	(4)	(5)
$\delta_f{}^{239}Pu/\delta_f{}^{235}U$	$1.284 \pm 4\%$	1.28	1.216	1.211	1.215	1.210	1.193
$\delta_f{}^{238}U/\delta_f{}^{235}U$	$0.0789 \pm 4\%$	0.0745	0.0847	0.0841	0.0853	0.0838	0.0815
$\delta_f{}^{237}Np/\delta_f{}^{235}U$	$0.542 \pm 5.5\%$						0.436
$\delta_c{}^{238}U/\delta_c{}^{238}U$		1.44	1.334				

*Experimental error is standard deviation.
(1) Rapsodie 64-2, Hansen and Roach set, 10 groups, 2D diffusion.
(2) Rapsodie 67-1, Hansen and Roach set, 10 groups, 1D diffusion.
(3) Rapsodie 67-1, Hansen and Roach set, 10 groups, DSN transport.
(4) Rapsodie 67-1, Hansen and Roach set, 16 groups, diffusion and correction for dilution.
(5) Rapsodie 67-1, Russian ABBN set, 25 groups, correction for dilution.

Table 3   Equivalent Flux at Core Center[3]

Detector	Flux at core center at 20 MW(t), neutrons/(cm²)(sec)	
	Calculated*	Measured
Ni	8.06×10^{14}	6.6×10^{14} (flux for equivalent fission)
Rh	1.2×10^{15}	9.3×10^{14} (flux for equivalent fission)
Au	2.22×10^{11}	1.25×10^{12} (flux per unit lethargy at 4.9 eV)
Mn	1.34×10^{12}	1.26×10^{12} (equivalent thermal flux)

*NIOBE code with 52 energy groups.

Table 4   Gamma Heating Rates in Core Central Channel and in
11th-Row Experiment Channel[4]

Experiment position and date	Distance from core midplane, cm	Sample	Gamma heating rate at 8 MW, W/g		
Central channel, 2/7/69	+40	Stainless steel	0.06		
	+16	Stainless steel	1.47		
	0	Stainless steel	2.32		
	−15	Stainless steel	1.24		
	−35	Stainless steel	0.09		
			Gamma heating rate, W/g		
			At 6 MW	At 12 MW	At 24 MW
Channel in the 11th row, 12/14/67	+40	Graphite	0.014	0.020	0.016*
	+20	Stainless steel	0.013	0.022	0.044
	0	Graphite	0.030	0.061	0.095
	−20	Stainless steel	0.012	0.024	0.046
	−40	Graphite	0.011	0.024	0.040

*Calorimeter functioned abnormally.
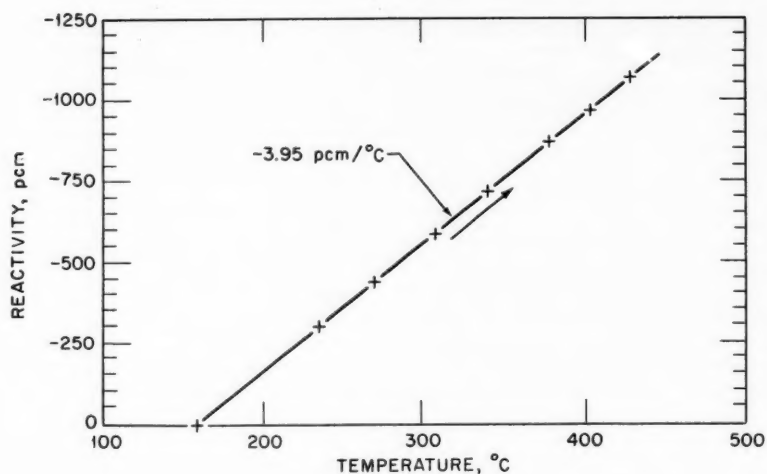
Fig. 12 Isothermal temperature coefficient[5] of reactivity (56 subassemblies). One dollar is approximately 532 pcm for Rapsodie.
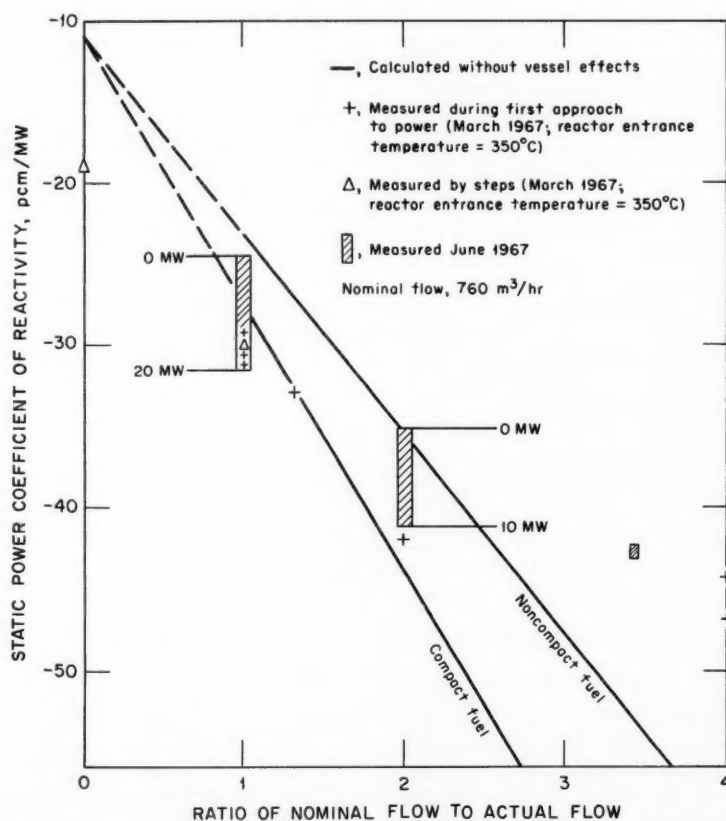
Fig. 13 Static power coefficient[5] as a function of coolant flow.
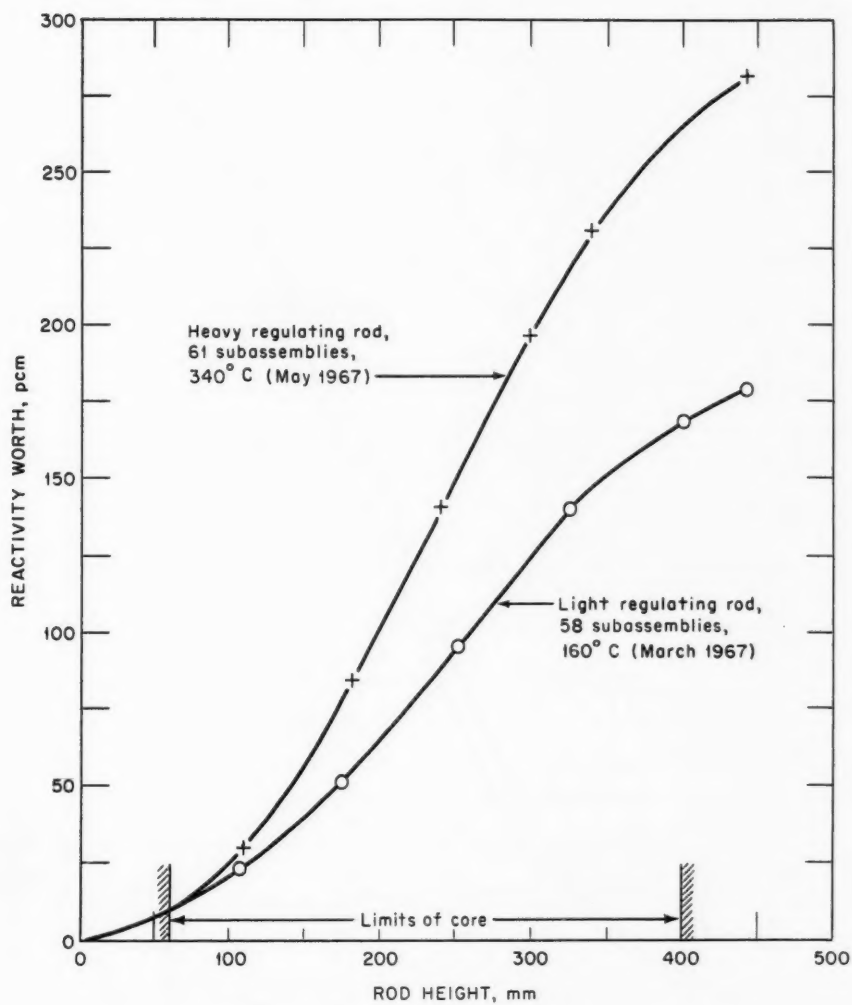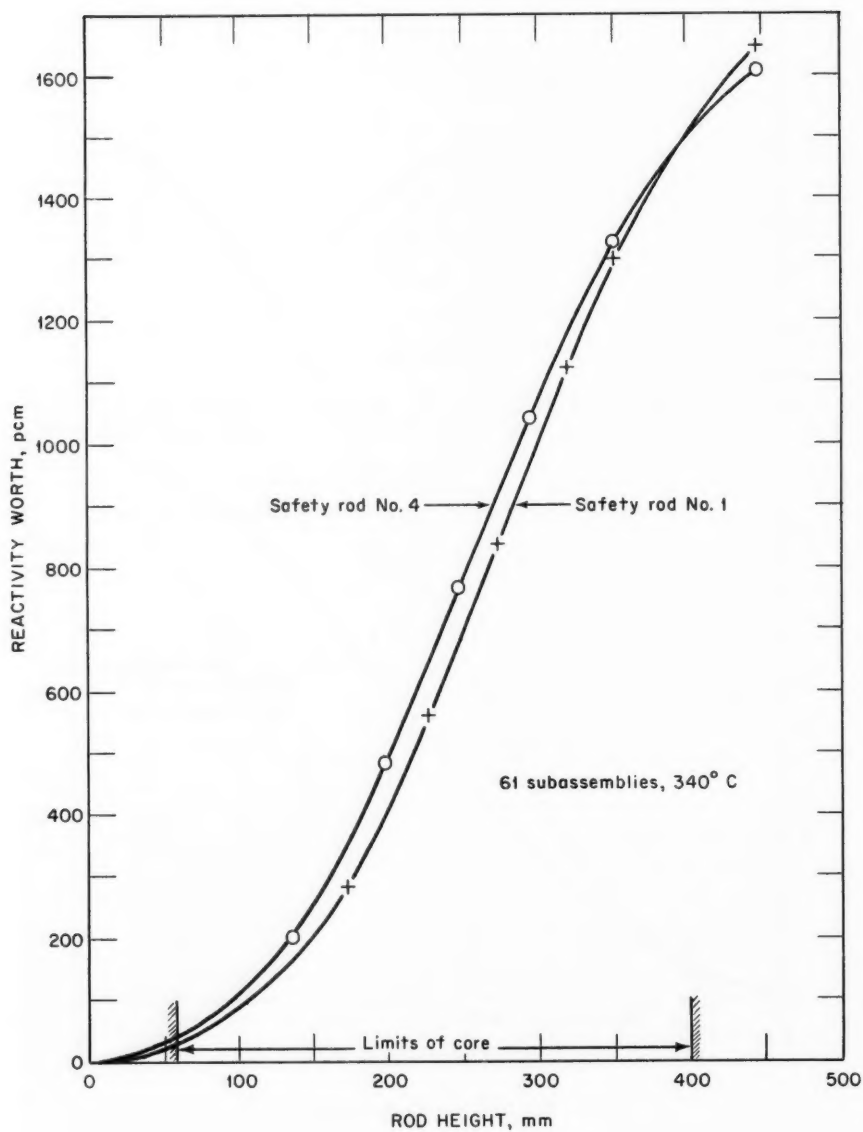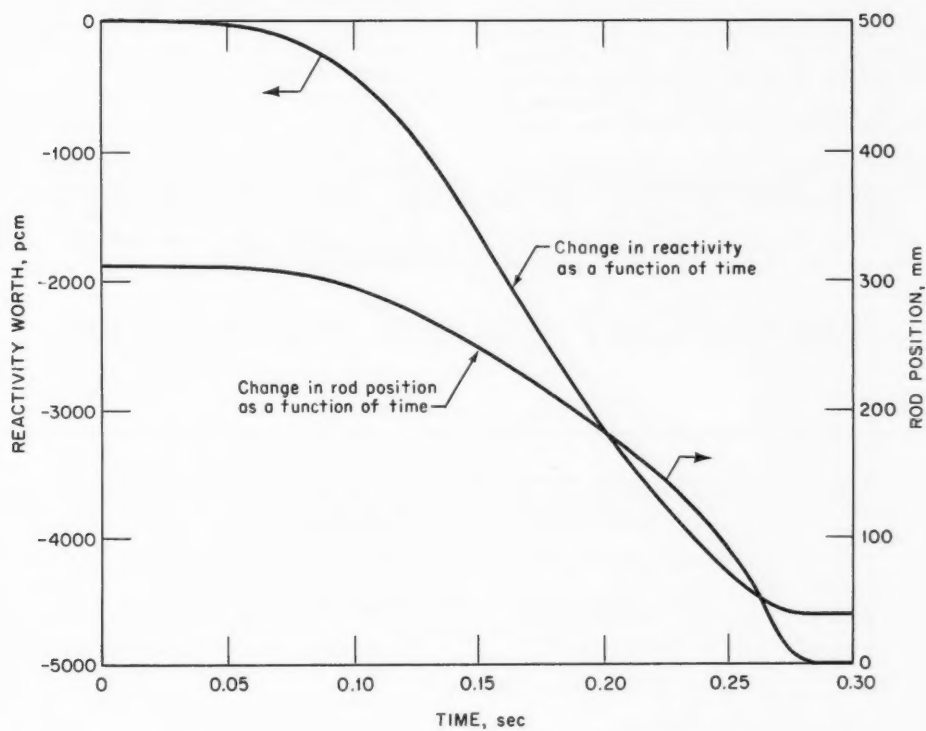
Fig. 14 Reactivity worths of regulating rods.[5]

Fig. 15 Reactivity worths of safety rods.[5]

Fig. 16   Rod-drop measurements[5] for control-rod bank.

Table 5   Summary and Comparison of Rod-Drop Measurements[5]

Methods	Safety rod No. 1 full-length drop		Rod bank from simulated 20-MW level	
	Δk, pcm	Drop time,* sec	Δk, pcm	Drop time,* sec
Direct measurement†	1641	0.32	5480	0.24−0.27
Rod drop				
By extrapolation	1730		5160	
By "inversion"	1540		4590	0.28
Reactivity meter	1860			

*From the beginning of rod motion to total stopping, not accounting for circuitry dead time.

†Static reactivity calibration; direct readout of drop time.

REFERENCES

In view of the significance of the material on Rapsodie, I have deviated from the established practice of using reference material that is available from the National Technical Information Service as stated on the inside front cover. The references listed below may instead be purchased (in French) from Direction de la Documentation, 31 quai Voltaire, Paris VIIéme.

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FOURTH CONFERENCE ON PLASMA PHYSICS AND CONTROLLED NUCLEAR FUSION RESEARCH

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Interested people in the United States can obtain further information from John H. Kane, Special Assistant for Conferences, Division of Technical Information, U. S. Atomic Energy Commission, Washington, D. C. 20545.

Summary of Second Symposium on Developments in Irradiation Testing Technology

By Myrna L. Steele*

The second symposium on irradiation-technology developments was held at the National Aeronautics and Space Administration's Plum Brook Station, Sandusky, Ohio, on Sept. 9–11, 1969. Brief summaries of the papers by session are given in this article. The full papers have been published in the conference proceedings, which are available for $3.00 from the National Technical Information Service as report CONF-690910. Sessions included: capsule designs and experimental techniques for fast reactor irradiations; irradiation-environment control; irradiation-environment determination; postirradiation examination; general irradiation techniques and applications; and capsule design, fabrication, and operating experience.

SESSION I: CAPSULE DESIGN AND EXPERIMENTAL TECHNIQUES FOR FAST REACTOR IRRADIATIONS

Citing the need for more accurate cross-section data for the (n,α) reaction for 304 and 316 stainless steels, Moteff[1] used the reported data on fast-neutron spectra and on (n,α) cross section vs. energy to determine the spectrum-averaged cross sections for five reactions: $^{54}Fe(n,\alpha)^{51}Cr$, $^{14}N(n,\alpha)^{11}B$, $^{27}Al(n,\alpha)^{24}Na$, $^{59}Co(n,\alpha)^{56}Mn$, and $^{93}Nb(n,\alpha)^{90}Y$. He then computed the quantity of radiation-induced helium atoms generated in the 304 and 316 stainless steels for the various irradiation environments that exist in the Engineering Test Reactor (ETR), the Dounreay Fast Reactor (DFR), and the Experimental Breeder Reactor II (EBR-II). Moteff proposed some corrections to the fission-spectrum-averaged cross sections to provide semiquantitative calculational methods for determination of amounts of helium generation under irradiation conditions in reactor environments.

Beeston, Ellingford, and Brinkman[2] covered experiences in design and fabrication of fatigue-specimen capsules in special EBR-II irradiation subassemblies. They assessed the effects of program criteria on capsule design, summarized the operating experience with irradiation subassemblies, and discussed the types of temperature monitors and the EBR-II fast reactor irradiation environment. Robinson, Ivins, and Dickerman[3] also discussed the use of a liquid-metal-cooled irradiation capsule for LMFBR-oriented materials and safety studies. Their paper centered on capsules that were designed for the simulation of various fast reactor accident conditions in a sodium-cooled thermal-reactor test-loop environment. The authors traced the evolution of irradiation capsules for use in the Transient Reactor Test (TREAT) facility from the original graphite-lined stainless-steel capsules (for static water-reactor fuel-melting experiments) through development of a transparent capsule for use with a high-speed color-photography capability. The use of both water and liquid-sodium autoclaves and integral sodium test loops was outlined. The authors then detailed the conditions under which both the first- and second-generation integral sodium loops and the high-pressure sodium autoclave operate.

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Dickerman et al.[4] summarized the TREAT-design goals and parameters. Some facilities unique to the TREAT installation were described; these are: the viewing slot that extends to the core center, the fast-neutron hodoscope system, and the neutron radiographic facility. The maximum transient conducted in TREAT, under the operating limit of a temperature-limited transient at 600°C, produced a maximum power of 1.7×10^5 MW and an integrated power at 60 sec of 2.15×10^3 MW-sec for an input of 4.9% k. The authors then summarized the various testing programs for which TREAT has been used.

Walker, Halfman, and Sanecki[5] discussed the methods of xenon tagging of instrumented subassemblies that were designed for use in the EBR-II. They compared tabularly the isotopic compositions of natural xenon and the special xenon-tag gas used in the elements that comprised the instrumented subassemblies. The xenon-tagging equipment and its operation were described, and general procedures for the tagging process were listed. Also given were the xenon recovery and injection times for some of the instrumented-subassembly fuel elements. Hutter, Seim, and Smaardyk[6] described in detail, after their short description of the EBR-II, the instrumented-subassembly system and the in-core instrument-test facility of that reactor. These test facilities are intended to provide remote instrument-readout capability for data acquisition from the experiments while the reactor is operating. The operating conditions for the instrumented-subassembly system and the in-core instrument-test facility were discussed. The major components of the instrumented-subassembly system are the subassembly that contains the fueled capsule, the subassembly extension, the bellows-seal assembly, the subassembly-drive system, and the data-logging system. The authors also gave cursory descriptions of the components of the in-core instrument-test facility. Seim, Ware, and Hutter[7] covered the 16 types of irradiation subassemblies that were used in the EBR-II; the authors tabulated design parameters for the capsules or subassemblies and noted the unique features. Drawings illustrated the design details of the subassemblies.

Montgomery, Grando, and Strasser[8] discussed a design for an instrumented capsule for fuel-swelling and fission-gas-release experiments. The fuel specimens to be used in these capsules were described briefly, and capsule materials and operations data were tabulated. The capsule assemblies contain a binary gas system for temperature control and thermocouples for temperature measurement. The authors outlined the results of the unrestrained-swelling and fission-gas-release experiments.

Olsen, Fitts, and Cox[9] compared the neutronic and thermal environments for a stainless-steel-clad $(U,Pu)O_2$ fuel rod operating in a typical proposed commercial fast reactor with test conditions available in both thermal and fast-flux test reactors. The authors made a quantitative assessment of the neutron fluence at the fuel cladding, radial temperature and power distributions, fuel restructuring, and fission-product yield as a function of burnup. They outlined their basic assumptions and calculational procedures and discussed in some detail the results of these procedures. They presented graphically the results of central temperature calculations for fuel pins during irradiation and showed power, temperature, and flux profiles for several reactor environments and flux irradiation levels. Fission-product composition within the irradiated fuel was tabulated as a function of time. Calculational results for thermal and fast test reactors (e.g., ETR and EBR-II) were compared from a viewpoint of simulation of actual operating conditions for large, fast power reactors.

SESSION II: ENVIRONMENTAL CONTROL

Struss and Chen[10] described a test facility that provides a variable temperature in a fast-flux environment. This facility is installed in a side vertical thimble in the Ames Laboratory Research Reactor. Before installation of the flux converter, the measured thermal flux in the vertical thimble was 1.6×10^{13} neutrons/$(cm^2)(sec)$. After the flux converter was installed, the measured thermal flux in this thimble was 1.5×10^{13} neutrons/$(cm^2)(sec)$ with a cadmium ratio of about 5 for gold; the maximum fast flux was 6×10^{12} neutrons/$(cm^2)(sec)$ $(E > 1$ MeV). The authors described the design and fabrication of the cryostat system, which was designed on the thermosiphon principle of operation. They summarized the gamma-ray-heating and gamma-flux measurements in the flux converter and in the core.

Zielenbach and Miller[11] discussed a capsule-system design that had an annular heat pipe incorporated into the capsule to achieve a uniform temperature over the fuel-sample length. They discussed the mockup experiment that was designed to evaluate the heat-pipe irradiation-capsule concept; optically measured temperatures in the mockup were tabulated. The authors briefly analyzed the overall results from the mockup experiments. Robinson and Basham[12] also described a test capsule for measuring radiation effects on fuel

specimens. They discussed the measurements of in-pile compressive creep of ceramic fuel materials; the compressive load is applied to the specimen through a bellows-sealed gas-actuated piston. The authors listed the design criteria for the in-pile creep-measuring apparatus; each component of the test rig was then described. They outlined the results of mockup and laboratory prototype experiments; effects of temperature, compressive force, and radiation were shown graphically.

Weaver, Patrick, and Ranken[13] discussed the development of an isothermal regulator for self-regulated constant-temperature irradiation experiments. The irradiator contains a heat pipe that is filled with an inert gas in addition to the working fluid. The authors described the principle on which this heat-pipe irradiator works. Results of bench tests and tests in the Omega West Reactor were given, and temperature distributions were shown for both types of measurements. They briefly described the design for an instrumented isothermal irradiator for EBR-II radiation-effects experiments. Another design for a 30-kW circulating-helium test loop was described by McDonald et al.[14] The test rig was designed for fuel-element-specimen testing in the Plum Brook Reactor (PBR). The authors gave the detailed design characteristics and performance capability of the circulating-helium rig. They also discussed the performance of the loop during both in-pile and out-of-pile endurance testing.

Stein, Homyak, and DeFayette[15] discussed an experimental technique used to simulate a uranium nitride (UN) fuel pin. The mockup was effected by fully enriched uranium-metal sheets that were sandwiched between aluminum foil and rolled around a spindle. The authors described the methods used for axial and radial power-distribution measurements and the program for radiation-effects testing in NASA's 100 kW(t) Mock-up Reactor [MUR, a core mockup of the 60 MW(t) PBR]. Use of a hafnium shield for shaping the neutron flux inside the capsule was discussed, and the effects of the shield on power-density distributions were shown graphically. The authors analyzed MUR flux and power-density data from the viewpoint of their use for design of fuel-pin irradiations in the PBR.

Lee and Werner[16] described the design and operation of a gas-buffered annular-heat-pipe fuel-irradiation capsule that they developed for use in the General Electric Test Reactor (GETR). The requirements for capsule design were listed. The authors developed the equations for maximum heat transfer with considera-

tion for the capillary-pumping and pressure-drop constraints and then described the bench-model tests by which they verified these heat-transfer equations. They briefly evaluated the potential application of this gas-buffered heat-pipe capsule for irradiation of reactor fuels and materials.

SESSION III: ENVIRONMENTAL DETERMINATION

Power[17] described the use of oxalic acid (H_2Ox) for gamma-dose-rate measurements in the PBR core when the reactor is operating at 40 and 60 MW(t). He discussed the method by which the gamma radiation induces the decomposition of the H_2Ox and gave the detailed methods for carrying out the H_2Ox radiation and analyzing the off-gas products as a function of time. The design parameters for the experiment facility, which was used in the core, were tabulated. Power[17] graphically presented the gamma-heating values (watts per gram) as a function of distance above core midplane; also presented were the H_2Ox decomposition and the carbon dioxide-, hydrogen-, and oxygen-generation rates as a function of time after start of irradiation.

Folkrod[18] discussed the development of fission-gas pressure transducers at Argonne National Laboratory (ANL). He outlined the basic design and operation of the null-balance and fluid-filled bellows-capillary types of transducers and discussed the unique method that ANL developed for control of the null-balance transducer. He then summarized the results of calibration and performance testing of this transducer. The data were presented graphically. Problems encountered in the development of capillaries for the fluid-filled bellows-capillary type of transducer were also described by Folkrod. He showed graphically the effects of temperature and voids within the metal and included a typical calibration curve for this type of transducer.

Yarger, Miller, and Ray[19] described a capsule that was used for continuous monitoring of fission-gas release from fuel samples undergoing irradiation. This capsule system consisted of a null-balance pressure transducer, which was attached directly to a clad fuel specimen in the capsule, and instrumentation for monitoring pressure balance. The authors described the method for the capsule-system operation and summarized the results of operation of two different capsules. Burley[20] also described a method for measuring the internal-pressure buildup in full-sized mixed-oxide fuel rods during irradiation in the Pluto-

nium Recycle Test Reactor (PRTR). The tests were conducted on four instrumented Zircaloy-clad, vibrationally compacted, mixed-oxide fuel elements. Burley outlined the experimental procedures and described the instrumentation for the fuel rods. He then discussed the results of measurements of both the internal gas pressure and the plenum-gas temperatures and showed results graphically.

Schwarzer[21] discussed the development of high-temperature W—Re thermocouples. He described the method used to fabricate the thermocouples and detailed the high-temperature thermal cycling and drift testing; results of the tests were presented in graphic form. Schwarzer then discussed results of postirradiation analyses of the thermocouples, including visual inspection, chemical analyses, microprobe analyses, and metallographic examinations.

Salgado, Thamer, and Rudman[22] discussed the development of grooved melt wires for fuel-element temperature measurements in the Ultra High Temperature Reactor Experiment (UHTREX). The melt wires were developed for temperature measurements over the 250 to 3000°F range. The authors tabulated the performance data of the wires over this range by wire composition. They described the melt-wire technique and the methods of obtaining information by this procedure. In-core temperature measurements during reactor transients were described; temperature profiles for both steady-state and transient conditions in UHTREX were included.

Zimmerman[23] described the design of a calorimeter that was used to measure heat-generation rates of several materials in a reactor test-facility environment. He discussed the characterization of the reactor environment and the energy-deposition mechanisms. He then described the calorimeter and summarized the features of each component. Characteristics of the samples used in the energy-deposition measurements were listed, and the results of the measurements were tabulated. Zimmerman analyzed the results in detail and discussed the manner of separating the gamma and thermal-neutron contributions to the overall heat-generation rate.

Stillman and Chaney[24] reported on a miniature neutron-flux monitor for temperature—time history determinations in neutron populations having an e-folding time of 20 nsec. The principle on which the fission couple operates and the application of the principle were outlined. Effects of bead size and conductive heat properties were detailed. Use of the fission-couple monitor for neutron-spectrum and -flux mapping was discussed. The authors then evaluated the

fission-couple performance in a steady-state radiation environment.

The use of high-speed photography and a transparent autoclave for observing fuel-failure behavior was discussed by Barghusen et al.[25] They described the transparent autoclave and the fuel-element design. Methods of analysis of the high-speed film, experimental data, and fuel-failure behavior were outlined. The authors showed some results that were obtained by the transparent-autoclave—high-speed-photography technique.

SESSION IV: POSTIRRADIATION EXAMINATION

Sowa[26] described the basins used at Hanford for postirradiation underwater measurements of fuels and test capsules. He summarized the five methods of obtaining dimensional measurements: optical comparators, hydraulic gauging, ultrasonic transducers (nuclear fuels), strain-gauge transducers, and variable reluctance transducers (nuclear fuels). Dismuke[27] discussed three Oak Ridge National Laboratory techniques for obtaining postirradiation data on space reactor fuels. In the three techniques, irradiation capsules are punctured by a pycnometer, a profilometer, and a laser, respectively. For each technique, the author described the associated equipment, discussed the methods of using the equipment, and included schematics. Morgan and Towns[28] described the development of techniques and equipment for postirradiation handling of coated microspheres of nuclear fuel. They described the procedures for preparation, handling, and inspection of the irradiated coated particles and the equipment for carrying out the procedures.

Ball and Sheibley[29] discussed and evaluated a unique method for determining fuel burnup and fission-product release in high-temperature low-burnup W—UO_2 plates. In this method ^{184}W is the thermal-flux monitor and ^{137}Cs is the burnup monitor. The ^{137}Cs was obtained from leaching of the capsules. The authors evaluated the data for ^{131}I that resulted from the leaching methods, as well as the data for ^{131}I and ^{135}I releases on the basis of the number of fissions calculated from the flux values obtained with the ^{185}W dosimeter. Also discussed were the problems encountered when conventional methods were attempted for determination of fission-product releases and fuel burnup.

Daugherty, Lieberman, and Doumas[30] described their simplified techniques for calculating the decay-heat-power levels for irradiated fuel specimens. Con-

tributions to the total power from fission-product and activation-product decay were considered. To verify their calculations, the authors described experiments in which decay power was measured for irradiated UO_2 and ThO_2 fuels; a comparison of measured and calculated decay power was tabulated. They showed the total decay power and the gamma contribution to the total decay power as functions of time subsequent to irradiation. They next described experiments for heat-flux and heat-transfer measurements for 7- and 10-rod bundles and showed the results graphically.

Albrecht, Johnson, and Hayes[31] described a special postirradiation examination facility for analyzing fuel samples 0.20 in. in diameter by 0.75 in. long. They summarized the techniques and procedures used at the facility and presented the design characteristics of the apparatus for decladding, subdividing, and analyzing the irradiated fuel specimens.

SESSION V: GENERAL TECHNIQUES AND APPLICATIONS

Lieberman[32] discussed calculations of fission power in irradiation-test loops for fuel samples in the ETR. He showed graphically the effects of control rods and test-loop cooling-water conditions on test fission power. Empirical methods of determining fission power in a test loop were discussed, and equations were given. Lieberman compared the calculated values with the measured values.

Serpan and McElroy[33] described the application of the damage-function concept to the analysis of radiation effects as a function of temperature on a 6-in.-thick A-302B structural-steel plate. They discussed the mechanical properties of the steel plate and included Charpy V-notch ductility characteristics, transition temperatures, and neutron-fluence data for irradiations at 450°F. Damage-function equations were formulated, and the solutions to these equations were obtained. The authors applied this procedure in an analysis of radiation—temperature effects on the 6-in. plate and tabulated the resulting group-averaged values of the damage function. Then they compared the measured total fluence with the calculated total fluence. They also tabulated the calculated predictions of fluence and energy response necessary to produce a 200°F ductile—brittle transition-temperature increase in A-302B steel for various reactor environments (e.g., Big Rock Point, CVTR, and EBR-II).

Carter[34] discussed the use of underground nuclear explosions as neutron sources for determining radiation effects on materials. He characterized the energy density associated with such intense sources and gave working equations for obtaining values of electromagnetic radiation arriving at the aboveground experiment location, together with equations for beam intensity, neutron energy, and neutron flux. He described the physical problems associated with the use of underground explosions for experimental purposes. Carter summarized his radiation-annealing experiments and considered the effects of high-intensity-radiation damage on these experiments.

Zelezny, Gibson, and Graber[35] discussed microprobe techniques and applications for determining the relative degree of fission-gas retention by irradiated dispersion fuel elements. They described sample preparation and showed some results of microprobe scans and photomicrographs. The authors then described the shielding and instrumentation that were used in the shielded-microprobe analyses. They compared the fission-gas-retention ability of irradiated oxide fuel with that of irradiated intermetallic-dispersion fuel. Ray[36] considered the use of neutron radiography as an analytical tool in irradiation-testing technology and described the neutron-radiography facilities at Battelle Memorial Institute. He discussed the determination of specimen dimensions, showed fuel-specimen swelling as a function of burnup, and noted briefly the use of neutron radiography for examining the condition of a test piece before the piece is inserted in a reactor. Hillig[37] also discussed the use of neutron-radiography techniques for inspection of irradiated fuel. He reported that the technique was applied to SNAP fuel elements and that the results were correlated with hot-cell data obtained by destructive analysis. Hillig described the neutron-radiography and fuel-handling facility at Atomics International. He discussed the application of neutron radiography to fuel swelling, cracking, and decomposition and to the quantitative determination of the hydrogen content in U—ZrH fuel elements and of the gap between fuel and cladding.

Sandefur et al.[38] discussed the results of extensive in-pile performance tests on Chromel—Alumel and W—Re sheath-type thermocouples. Fabrication data for the thermocouples were tabulated, and construction of capsules for in-pile thermocouple testing was described. The detailed data from the irradiations were tabulated, and such parameters as operation time, neutron fluence, junction type, and average and maximum temperatures were included. The authors analyzed the data and extracted thermocouple-reliability factors. They evaluated the effects of thermocouple location in the core, fast-neutron fluence (defined as $E > 0.18$ MeV), and temperatures; they also showed graphically

the percentage of thermocouple failure as a function of operation time. Finally, the authors summarized the types of thermocouple failure modes.

Buck[39] described the conceptual Continuous Remote Instrumentation and Core Evaluation Technique (CRICET), which is a system for remote measurement of fuel-element temperatures. He included design and function diagrams and evaluated the effects of reactor environment on in-core instrumentation. The functioning, operation, and accuracy of the individual subsystems were discussed, as were the important features of CRICET.

Albrecht et al.[40] discussed irradiation studies of uranium nitride (UN). They described capsule design and fuel-specimen fabrication and cladding. Specimen temperature calculations and the results of postirradiation examinations of samples were tabulated. The authors reported microscopic and macroscopic observations and included neutron radiographs showing dimensional changes in the samples. Chemical techniques for burnup analyses were discussed. Analyses of the mechanisms of UN swelling were compared with analyses obtained in the SNAP-50 program.

SESSION VI: CAPSULE DESIGN, FABRICATION, AND OPERATING EXPERIENCE

Reilly[41] described the operating experience on in-pile irradiation of tungsten-clad uranium dioxide (UO$_2$) fuel specimens. He discussed methods of predicting temperatures in the fuel forms and considered problems related to measurement of fuel-cladding temperatures in the 1800°C range. Reilly[41] briefly discussed the uses of MUR irradiations prior to long-term irradiation in the PBR.

Fromm and Miller[42] discussed experiments in the MTR (Materials Testing Reactor), ETR, and EBR-II for irradiation of mixed (U,Pu)N fuels. They described the capsules that were designed for the MTR—ETR irradiations and the results of the heat-generation measurements. The authors then discussed the basis for setting up the EBR-II irradiation program. They described the capsule-assembly program, the sodium bonding, and the eddy-current testing.

Heckelman[43] described the design of a retractable—replaceable thermocouple for use in in-pile instrumented-fuel-capsule experiments. He provided a detailed table showing thermocouple characteristics, the irradiation environment under which the thermocouple must function, and thermocouple performance. He discussed the design objectives and listed the basic

auxiliary equipment required for this thermocouple. Heckelman then summarized the in-pile and out-of-pile performance tests and gave diagrams to show step-by-step replacement techniques for a typical thermocouple that was being irradiated in the core.

Tunnell and Brown[44] discussed the development of a facility for fuel-irradiation tests in a flowing-liquid-metal environment. They briefly described the capsule design and the test environment in the GETR. They discussed the results of thermal analyses and included the equations used to evaluate the heat-transfer characteristics of the capsules.

Novak et al.[45] discussed the irradiation testing of fast reactor fuels. They described the operation of a forced-convection capsule for determining stoichiometric effects on fuel swelling and/or fuel-erosion characteristics of defected mixed-oxide fuel under flowing-sodium conditions. The authors evaluated the performance of the electromagnetic sodium pump and tabulated performance characteristics.

Sheibley[46] discussed a design for a lead-type irradiation capsule for experiments on radiolytic decomposition of water in aluminum capsules. He described the experiment system and capsule and discussed the reactor irradiation environment for these. He summarized the system capabilities for experiment execution. The theoretical basis of this experiment is then discussed by Sheibley, Holms, and Sidik.[47] The design technique, called "doubly telescoping," accounts for control-rod and reactor-cycle block effects and the interaction of the multiple effects of the reactor environment. The authors defined a multivariable polynomial equation that accounts for the n-order interactions; they discussed and defined the theoretical bases for orthogonally blocked experiments and telescoping sequences. They then illustrated the application of theory to experiment by a sample problem.

Montgomery, Grando, and Strasser[48] discussed the results of fast reactor fuel irradiation in a simulated fast reactor environment in a thermal reactor. The testing of sodium-bonded and helium-bonded fuels was compared for thermal vs. fast test reactors. The authors described the plutonium-containing fuel capsules for irradiation in the GETR. They considered the problems associated with using a sodium bond as the temperature increases during irradiation.

Jacobson, Kerkhoff, and Christenson[49] described the design of a capsule for dynamic irradiation testing of solid lubricants to evaluate the effects of fast-neutron flux and fast-neutron fluence. They also described the in-pile friction-and-wear testing appa-

ratus. The reactor environmental conditions were summarized, and the proposed in-pile operational modes were outlined.

Culley and McMahan[50] discussed the equipment and procedures for remote encapsulation of preirradiated fuel pins for irradiations in TREAT. They showed and described a capsule and its assembly, the flow diagram for capsule loading, the profilometer and its associated instrumentation, the gamma scanner and its associated electronics, the remote welding fixture, the NaK-fill system, and the cask-transfer and -positioning system.

Rohal[51] described the design of an irradiation capsule and commutator assembly for continuous-capsule-data readout without having leads attached directly to the capsule. The commutator assembly is installed in the reactor core at the normal end-stop position so that, when the capsule is in the fully inserted position, electrical contact is made between the capsule and the commutator assembly. The capsule lead pins are electrically insulated from the capsule body and are attached to sensors within the capsule. The end-stop commutator is connected by permanent lead to a recorder. The special problems involved with this type of design were discussed.

ACKNOWLEDGMENT

The author wishes to thank Stanley Goldsmith of Battelle—Northwest and Technical Chairman of this symposium for his suggestions and his review of the manuscript.

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FOR *REACTOR TECHNOLOGY*

One interpretation of the words "Critical Review" emphasizes the word *critical*. That type of article would have as its purpose the discussion of a single subject, which has been uncertain and perhaps controversial, in considerable depth. An example might be a critical review of the values of alpha for plutonium, assessing all the work done and arriving at a best current estimate. This paper is primarily addressed to specialists on the particular subject, and serves as an authoritative source for the information they use.

In the second interpretation of a Critical Review, the emphasis is on the word *review,* in the sense of survey. Such a paper would be broader and probably more descriptive in scope. A paper on "Solubility of Metallic Elements in Liquid Sodium" is an example of this category. Such a paper would be addressed to a much broader group of readers and written in a fashion that would be of interest to the majority of reactor technologists. It would provide enough of an introduction that the specialist from another field could immediately appreciate why the subject is important.

The criteria for these two types of articles may not be very different:

1. The paper should be based on a thorough coverage of relevant work on the subject from many sources. A review based on the work of only one individual or group is better suited for publication in one of the regular society journals. The review should be critically selective, reporting only the most valid results and indicating why some prior results have a questionable status. The review should call attention to significant gaps where more work is required in the subject of the article.

2. The paper should be timely, on a subject of active current interest.

3. The scope of articles acceptable as Critical Reviews includes all the subject areas identified for *Reactor Technology* (Economics, Physics, Mechanics, Construction, Fuel Elements, Fuel Cycles, Fluid and Thermal Technology, Fuel Processing, Components, Operating Performance), as well as Materials (including Source and Special Nuclear), Environmental Effects, and Effluent Management. Not desired are (1) reports of original research proposed for first publication and (2) review articles directed toward the specialist in the field of nuclear safety (which is covered by the AEC's bimonthly review *Nuclear Safety*).

4. The paper should be organized so that it is of immediate practical use to the readers. Such organization requires: attention to consistent use of units, presentation of important data in summary curves or tables, and a fully adequate bibliography to the original literature. Details on size, honorarium, style, etc., can be obtained from ANS.

Status of Reactor-Physics Calculations for U. S. Power Reactors

By Jack Chernick*

HISTORY OF REACTOR-PHYSICS CODES

Reactor physics evolved during the Manhattan Project of the 1940s on the basis of simple physics experiments and theoretical methods devised by Fermi, Wigner, and other notable physicists. Today there is an almost complete dependence on the modern electronic computer, on programmers, and on codes with a profusion of names. Many links with the past are fading, especially since much of the record of the Manhattan Project now exists only in the form of a few rare copies of papers and handbooks.[1,2]

In the early years after World War II, reactor-physics calculations were based principally on the 4-factor formula, one or two neutron-energy groups, and on diffusion or age-diffusion theory. The rise of more detailed multigroup methods, first attempted by F. L. Friedman of the Manhattan Project, became feasible only with the development of high-speed digital computers. The speed and memory of the latter machines increased rapidly, progressing through such stages as the IBM-650 and the UNIVAC to modern CDC-6600 and IBM-360 scientific computers. National laboratories built their own machines until the IBM-704 made them obsolete.

The naval reactors laboratories pioneered in the use of the electronic computer. At Knolls Atomic Power Laboratory (KAPL), motivation was provided by the complex spectrum of intermediate-energy power reactors which were then under study.[3] At Bettis Atomic Power Laboratory, the MUFT–SOFOCATE cross-section averaging codes and the PDQ

multigroup codes were developed for water reactors.[4] Modern descendants of these codes, such as LEOPARD,[5] are in wide use in industry.

The methods and formulas developed by the Manhattan Project formed an adequate basis for the design of well-thermalized graphite[6] or heavy-water reactors.[7] In fact, use of the 4-factor formula for D_2O-moderated reactors has persisted until recent times, not only in Europe and Canada but also in the United States. Honeck and Crandall[8] have shown that modern methods work equally well and permit a more detailed physical understanding.

The inadequacy of diffusion theory in predicting thermal-flux distributions within reactor lattices[7] first aroused interest in the spherical-harmonics methods developed by the Montreal group of Placzek, Marshak, and others.[9,10] For tight-packed water lattices, however, high-order P_n calculations or other transport-theory methods were needed for accuracy, and one-dimensional boundary conditions were found to be inadequate for rodded systems.[11] As more knowledge of neutron-thermalization processes became available, multigroup collision probability or integral transport methods were developed, such as THERMOS.[12] However, more approximate neutron-thermalization models are still in wide use.[4,5]

At Los Alamos Scientific Laboratory (LASL), two developments of future importance occurred toward the end of World War II. The first of these developments was the use of Monte Carlo methods in high-speed computers for criticality calculations. The second was the use of discrete ordinate methods for solving the neutron-transport equation. The latter work led to Carlson's S_n method,[13] descendants of which include, for example, the General Electric Company's

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SN-1D code[14] and the popular Oak Ridge National Laboratory (ORNL) 1D and 2D codes ANISN[15] and DOT.[16]

At the Brookhaven National Laboratory (BNL), early interest in slightly enriched uranium—water reactors led to a continuing program on the reactor physics of these lattices[17] which eventually was extended to ^{233}U and plutonium fuels as well. Manhattan Project methods proved inadequate for accurate treatment of neutron slowing down, fast fission, resonance absorption, and the thermal utilization of these lattices. One tool developed for treatment of the latter problems, the so-called collision-probability method,[11,18] has proved very useful because of its physical appeal and ease of computation. The method is still in wide use in studies of the resonance absorption of neutrons and neutron thermalization in reactor lattices.[19,20] Resonance-absorption codes based on collision-probability methods include ZUT-TUZ,[21] GAROL,[22] TRIXIE,[23] RABBLE,[24] and many others. THERMOS is widely used in such industrial reactor-design code packages as LASER[25] at Westinghouse Electric Corporation and CEPAK at Combustion Engineering, Inc.

The use of Monte Carlo methods for resonance-absorption calculations was first proposed by Richtmyer.[26] The resulting UNIVAC code REP proved very useful for tests of analytical methods that were then under development. Its limitations were gradually removed as improved codes, such as REPETITIOUS[27] and RECAP and its extensions,[28] came into existence. The most recent and most physically realistic of REP's descendants is REPCDC, programmed for the CDC-6600 computer.[29]

The use of Monte Carlo for fast-fission calculations was first made by Rief. The MOCA code[30] was improved and extended by Rief and others.[31] It is very useful in determining effects of reactor-lattice anisotropy, fast-advantage factors, fast fission, and moments of the neutron slowing-down distribution in reactor lattices.

With the increased speed of the modern computer, Monte Carlo codes, although expensive to run, are occasionally used by industry to check the accuracy of their standard reactor codes. Efforts to improve the speed, accuracy, and flexibility of Monte Carlo codes for particle transport continue at several laboratories, including ORNL, Pacific Northwest Laboratory, and LASL, and at Atomics International under the liquid-metal-cooled fast breeder reactor (LMFBR) program.[32]

Recommended for details on the computing methods used in reactor physics, a recent book[33]

contains chapters on multidimensional diffusion theory by A. Hassitt, the method of discrete ordinates by B. G. Carlson and K. D. Lathrop, spherical-harmonics methods by E. M. Gelbard, and Monte Carlo methods by M. H. Kalos et al.

We have sketched briefly the evolution of some of the codes used in reactor-physics design. Thus far we have said nothing about the many additional codes needed for studies of reactor-plant dynamics, control, and safety, shielding studies, fuel-depletion or fuel-management studies, and the LMFBR. We have said nothing about nuclear data or integral experiment data needed as input into these codes and little about the theoretical developments that underlie the present codes or point out the directions for future improvements in accuracy or costs. We shall discuss some of these matters, as well as current industrial practice, in the ensuing paragraphs. The areas selected for discussion reflect the personal viewpoint of the author, and no claim for completeness is made in either the discussions or references.

NUCLEAR DATA FOR REACTORS AND THEIR ANALYSIS

Today the measurement, compilation, evaluation, and use of nuclear data is a far cry from the few crucial cross sections and integral data measured or estimated during the Manhattan Project. After World War II the rapid progress in the construction of reactors, accelerators, and sophisticated instrumentation led to a trickle and then a flood of cross-section data. Under Hughes the compilation of neutron cross sections culminated in the BNL-325 and BNL-400 publications,[34] that are indispensable to experimentalist and reactor designer alike. The need for automating the storage and transmittal of measured data led to the magnetic-tape data library known as SCISRS,[35] which currently contains over a million data points. The four nuclear data centers at Brookhaven, Saclay, Vienna, and Obninsk together supply the world's needs for reactor cross-section data.

By mutual agreement a second-generation version of SCISRS is being prepared by the National Neutron Cross-Section Center (NNCSC) at Brookhaven. Similarly CINDA, an index to the literature on microscopic neutron data, first developed by H. Goldstein, is being kept up to date by cooperation among the U. S. Atomic Energy Commission (AEC) and nuclear data centers abroad.

The comparable automated library for evaluated data is the ENDF file developed by Honeck.[36] The

ENDF/B forms a library composed of a single (best) set of cross sections for reactor-physics codes in a continuing national program supported by the AEC.[37] The links in the chain between SCISRS, ENDF/B, and commonly used reactor-physics codes such as MUFT and GAM[38] (for thermal reactors) and MC[2] (for fast reactors)[39] have also been forged. These include the cross-section group-averaging codes ETOM (ENDF/B to MUFT), ETOG (ENDF/B to GAM), and ETOE (ENDF/B to MC[2]). The link between ENDF/B and THERMOS is accomplished via FLANGE, which is based on up-to-date neutron-thermalization data.

The data required for ENDF/B are evaluated by a large working committee, the Cross Section Evaluation Working Group (CSEWG), composed of representatives of various laboratories and AEC contractors and managed by the NNCSC at Brookhaven. Since CSEWG's organization in 1966, the ENDF/B Version I tape has been completed, checked for internal consistency, and tested against various integral data. Included are single-set data for about 80 isotopes for all important neutron-induced reactions from 10^{-5} eV to 20 MeV, as well as thermal-scattering-law data and cross sections for photon production and interaction. Allied areas are covered by the Radiation Shielding Information Center and the Nuclear Data Group at ORNL. Abstracts of computer codes[40] are maintained at the Argonne National Laboratory (ANL) Code Center.

Thermal-scattering-law data[41] for energies up to a few eV exist for a number of important moderators, including H_2O, D_2O, graphite, beryllium and its oxide, zirconium hydride, and a few organic moderators. The FLANGE II code[42] casts the pointwise ENDF/B data into multigroup form. At the Savannah River Laboratory, the latter have been checked where possible against such integral data as total cross sections, $\bar{\mu}(E)$, the average cosine of the scattering angle, neutron diffusion, and cooling coefficients.

The bulk of the experimental and theoretical work underlying the latter accomplishments was carried out during what may, in retrospect, be called the golden decade of reactor development, between the mid-50s, when secrecy wraps were first discarded, and the mid-60s, when reductions in AEC support of thermal-reactor development started. For lack of space we can name only some of the principal contributors to this work: Wigner, Wilkins, Radkowsky, E. R. Cohen, Brown and St. John, von Dardel, Van Hove, Zemach and Glauber, Krieger, Nelkin, Amouyal and Benoist, Takahashi, Honeck, Yoshimori and Kitano, Schofield, Vineyard, Corngold, and Michael, Young, and Koppel

on the theoretical side; Poole, Mostovoi, Egelstaff, Brockhouse, Haywood, Brugger, and Beyster on the experimental side. Their work is reviewed in a number of international conferences[20,43-46], including the Geneva conferences of 1955, 1958, and 1964.

Integral data such as neutron ages, resonance integrals, and measurements made in critical assemblies are very important in checking the reliability of cross-section data. Throughout the years large discrepancies have continued to occur between differential cross-section measurements and integral data. A few examples of historical interest include such integral data as the temperature coefficient of graphite lattices, which led first to more accurate measurements of low-energy values of eta for ^{235}U and later to discovery of the positive contribution of plutonium buildup to the temperature coefficient of Magnox reactors; the neutron age in water, which motivated more accurate measurements and calculations of neutron ages as well as relevant cross sections; the thermal value of eta for ^{233}U, which is so important for thermal breeders and which has remained remarkably stable despite alarms caused by discrepant (and later found erroneous) differential measurements; the epithermal $1/E$-integrated capture-to-fission ratio of ^{235}U; and many others.[47]

More recently a similar alarm was raised in the LMFBR program when British experiments[48] indicated that the values of α_{49}, the neutron capture-to-fission ratio in ^{239}Pu, in the 1- to 20-keV region were much higher than the U. S. evaluated data[49] contained in ENDF/B Version I. Further experiments both in England and in the United States have succeeded in more adequately pinning these numbers down in the 1- to 20-keV range between these two extremes.[50] The strong structural effects in alpha shown in the original British work still persists, however, indicating the complexity of the fission barrier[51] in ^{239}Pu.

The general adequacy of current Gwin–Schomberg data at energies below 1 keV has been confirmed by the analysis of BNL plutonium heavy-water lattices. Integral experiments in ANL's zero-power reactors have provided verification of the higher differential values for plutonium alpha in the 1- to 20-keV range. At energies above 20 keV, there is more uncertainty, as indicated by European work on the analysis of fast critical experiments.

The need for cross-section measurers, evaluators, and reactor analysts to interact more effectively has been recognized for some years. Several large conferences have been arranged for this purpose.[37,52]

The cross-section evaluator has a number of codes available to check measured cross sections or to fill in gaps where experimental data do not exist. Thus the ABACUS-II code[53] is routinely used within the CSEWG program to calculate elastic-scattering angular distributions. The NEARREX code[54] is an optical-model code that computes energy-averaged total, elastic-scattering, and reaction cross sections, as well as inelastic-scattering cross sections. The OPTIC program[55] goes one step further and includes resonance contributions to the cross sections. The 2-PLUS is a deformed-nucleus optical-model code.[56]

In the resolved resonance range, measured cross sections are represented on modern tapes by use of either a fine mesh or single-level Breit—Wigner parameters. In regions between resonances, "smooth" cross sections are used where necessary to make up for deficiencies in the single-level fit. Where interaction between resonances is important, as in fissile nuclides, the smooth cross sections may turn negative, leading to difficulties in some processing codes and to questions concerning the accuracy of multigroup cross-section sets. The practicality of using multilevel parameters, which would more closely fit the physical situation, is being increasingly explored.

Despite the great improvement in recent measurements and calculations of neutron cross sections, large gaps remain. One of the weakest areas is still that of inelastic cross sections and inelastic-energy degradation in the MeV range.* Recently the Oak Ridge Van de Graaffs (including a 7.5-MeV tandem Van de Graaff) have been used for such measurements, the accuracy of which is limited by corrections for multiple scattering in the sample. Another area is neutron capture (and fission) below 300 keV for major fissile and fertile nuclides. As a classical example, capture in ^{238}U in the unresolved energy range has always been based on the statistics of uncertain resonance parameters and p-wave strength function. The new accelerator ORELA (Oak Ridge Electron Linear Accelerator) was planned for time-of-flight measurements in this energy region. A new accelerator at Argonne, FNG (Fast Neutron Generator) complements the ORNL program.

In the area of thermal-reactor critical experiments, there exists a large body of data for simple light-water,

heavy-water, and graphite-moderated assemblies with enriched-uranium fuels, carried out at many AEC and industrial laboratories. Convenient compilations have been published of many of these data for uniform D_2O-moderated lattices[8] and for H_2O-moderated lattices.[57] Additional data are reviewed in various reports from industrial laboratories[58-60] and international meetings.[61-63] The data for water-moderated, plutonium-fueled lattices are relatively sparser. The chief sources are BNWL (Hanford) data[64,65] and the SAXTON[66] and ESADA[67] critical experiments.

There are additional commercially funded experimental data that have not been released for publication. A series of BNL exponential experiments on Pu—Al rods in D_2O over a wide range in lattice pitch was recently completed.[68] For ^{233}U-fueled assemblies, the experimental data are even sparser. Current work on such assemblies is primarily in support of the Navy's seed-and-blanket, water-moderated ^{233}U breeder.

The analysis of criticality data and measured cell parameters of reactor lattices has been carried out at BNL for many years[8,69-71] to determine the accuracy of the cross sections and analytical tools used. The basic codes used were MUFT, THERMOS, and more recently HAMMER,[72] together with transport and Monte Carlo codes to check the adequacy of the cell calculations.

Recently these methods, supplemented by the Monte Carlo code REPCDC,[29] have been used to analyze a number of H_2O and D_2O lattices to check the adequacy of the ENDF/B Version I data file and the HAMMER, ZUT-TUZ, and RABBLE resonance-absorption codes.[73] The latter codes consistently overestimate resonance absorption in ^{235}U and ^{238}U by several percent. A similar result was obtained for ^{239}Pu and ^{240}Pu resonance integrals in the BNL Pu—Al rod lattices in heavy water.

The inaccuracies in ZUT-TUZ include the neglect of overlap effects between resonances of different nuclides. The RABBLE[24] uses approximate cell-boundary conditions. Comparison with earlier results for enriched-uranium lattices showed a reduction in eigenvalues due mainly to a lowering of eta in the near thermal range in ENDF/B. In turn, the latter could be traced to the inexplicably large value of 2.75 b taken for the 2200 m/sec capture cross section of ^{238}U and the use of the "eyeball" curves, rather than the measured data for α_{25} shown in BNL-325(2nd Ed.).[34]

Clearly this last question, which is important for modern power reactors, will not be satisfactorily

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*A. B. Smith, Neutron Inelastic Scattering by Time-of-Flight, in *Nuclear Research with Low-Energy Accelerators*, J. B. Marion and D. M. Van Patter (Eds.), Academic Press, Inc., New York, 1967.

resolved until the variation of η_{25} with energy below its 0.27-eV resonance and its normalization are accurately determined. It was also found that changes in the scattering cross sections of graphite and heavy water in ENDF/B Version I disagreed with accurate neutron-age measurements. These latter discrepancies have been corrected in Version II.

Similar analyses of U–H_2O lattices have been made at other laboratories, the most recent example being that carried out at Bettis.[74,75] Industrial laboratories also attempt to bring their cross-section files, codes, and recipes up to date by testing them against criticality data.[5,58,76] All industrial code packages include fuel-depletion calculations; hence relevant experimental data on operating reactors are very important. Such data were obtained on the Yankee pressurized-water reactor[77,78] at Rowe, Mass. (Yankee–Rowe), and on some boiling-water reactors.[79]

In view of the pressures to recycle the plutonium produced in water reactors, a number of studies of technical feasibility and costs have been carried out for power plants for both boiling-water reactors (BWRs) and pressurized-water reactors (PWRs); a plutonium-fueled core was cycled through the Materials Testing Reactor (MTR),[80] and other experiments on plutonium-bearing lattices have been completed.[81] The adequacy of ENDF/B data for plutonium has been tested against resonance integrals and criticality data.[82]

Many industrial reactor physicists feel that the data needed for calculations for plutonium-fueled lattices or for reactivity changes in water reactors with long core life ($\approx$ 30 MWd/kg of uranium) are still inadequate. Among the integral experiments suggested are resonance-absorption measurements in mixtures of the important fissile, fertile, and fission-product nuclides; high-accuracy, detailed critical experiments on water lattices fueled with varying fissile and fissile–fertile contents of plutonium-enriched UO_2 fuel rods of 0.4 to 0.5 in. in diameter; hot critical experiments; and longer burnup data from operating reactors on bulk fission-product poisoning. The Savannah River Laboratory has found discrepancies in burnup calculations for plutonium isotopes, and Combustion Engineering, Inc., and Westinghouse Electric Corporation have encountered difficulties in harmonizing results obtained in plutonium and UO_2-fueled lattices without arbitrary changes in basic cross sections.*

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*R. L. Hellens, Brookhaven National Laboratory, personal communication.

Requests for additional cross-section data depend on the needs of the user. For power reactors, better data on important individual fission products are needed, including fission-product yields from ^{235}U, ^{238}U, ^{239}Pu, and ^{241}Pu fission. Since the need for accurate energy balances in power reactors has become important, there are requests for better data on component energy yields from fission, including the kinetic energy of fission fragments, and on prompt beta, gamma, and delayed-gamma energy yields. Production laboratories stress the need for better data on prompt-neutron yields of odd-mass isotopes of curium, the energy-group structure of delayed neutrons in fission, and the precision of fission-product yields, particularly for transplutonium isotopes, as well as better thermal and resonance data for ^{236}Pu and ^{238}Pu and their precursors. Reduced uncertainties in ^{233}U, ^{235}U, and thorium data are stressed for ^{233}U-based thermal power-reactor programs. The aerospace program stresses component cross sections and burnable poisons, primarily resonance data for isotopes of Hf, Gd, Sm, Eu, and Dy. Shielders want data on higher energy neutron cross sections and on gamma-ray production, cross sections, and energies.

Although AEC support of thermal-reactor programs has greatly declined, except in the area of nuclear safety, it is safe to say there is a continuing need of such programs. Measurements of both differential and integral data have become more precise with recent improvements in neutron-source intensities, experimental techniques, and calculational methods. Measurements of neutron spectra, reaction rates, fission tracks, and flux distributions, together with radiochemical and mass-spectrometric analyses, yield a more comprehensive set of data than was possible in the past.

In the analysis of critical experiments, some industrial users tend to omit exponential assemblies or critical assemblies with high leakage rates. One reason given is that power reactors are so large that neutron-leakage effects are not too important. Small assemblies are indeed frequently sensitive to reflector effects and fail to reach an asymptotic spectrum throughout the core.[83] Nevertheless, industry is now designing water reactors with regions of differing fuel enrichment or even checkerboard patterns to improve power distributions. Thus changes in neutron spectrum must be expected near region interfaces.

Our experience has been that such problems can be resolved at the expense of additional computer time. In the analysis of BNL tight Pu–Al lattices, the

HAMMER calculations were supplemented by ANISN to calculate the radial activations measured by various spectrum-sensitive foils. The resulting energy-dependent bucklings gave much more consistent eigenvalues than traditional fits to a single Bessel function.[84]

In the analysis of plutonium criticals, it is also necessary to raise the energy limit of standard neutron-thermalization codes such as THERMOS in order to properly treat the giant 1-eV resonance in ^{240}Pu. This problem is now recognized and is generally being remedied. More generally, in high-buckling systems, epithermal or fast,[85] it is necessary to treat with care the high-energy region where neutron scattering is strongly anisotropic. Slowing-down moments higher than the second are important in determining neutron leakage, and the difference between heterogeneous and homogeneous calculations is not trivial. Transport-theory codes with P_3 scattering[86] or Monte Carlo calculations are needed for accurate calculations. Finally, despite many theoretical discussions, the effect of heterogeneity on thermal-neutron leakage is not firmly established. Of course, this is of lesser import in water-moderated reactors that have small diffusion lengths.

The work in fast reactor physics in the United States is carried out as tasks under the LMFBR program. This program aims at developing the technology of mixed-oxide and sodium-cooled breeders and includes the construction of the Fast Flux Test Facility (FFTF) for fuel studies. The program involves industrial groups at all stages of the work, including research and development (R&D) and the design of target-size [1000-MW(e)] breeders, and encourages proposals for the construction of demonstration plants. By contrast, the development of breeder reactors abroad follows more traditional lines, emphasizes more flexible R&D programs and large-scale physics and fuel-cycle tests, and aims at the early completion of fast breeder prototypes. An excellent review of world progress in fast breeder development through 1968 is available.[87]

In the area of fast reactor physics, the LMFBR program stresses the measurement of high-priority cross sections, particularly at ORNL, ANL, and Rensselaer Polytechnic Institute (RPI), and cross-section evaluation work by the NNCSC at BNL and by CSEWG members at various contractor sites. Other cross-section measurements are supported by the AEC Research Division. A large-scale program of critical experiments is being carried out by Argonne. Other integral experiments, including spectra in material

blocks at RPI, blanket studies at Massachusetts Institute of Technology, Doppler and heterogeneity studies at Atomics International (AI) and Doppler studies by General Electric Company at the SEFOR reactor, are continuing. Work at other laboratories,[88] including BNL, ORNL, Gulf General Atomic, Pacific Northwest Laboratory, and Idaho Nuclear Corp., which have contributed to the integral experiment program, has recently been greatly curtailed.

The LMFBR program also supports large-scale code-package development by various contractors with emphasis on ease of conversion of these codes at different computer facilities. An attempt is being made to provide common input—output interfaces for such widely used codes[89] as MC^2, ANISN, DOT, and 2DB. Work is continuing on 3D codes, on Monte Carlo codes, on such synthesis codes as BISYN[90] and others, on such fuel-burnup and -management codes as FARED[91] and extensions of CITATION[92] (for the IBM-360 computer with over 200,000 words of memory), and on space-dependent reactor-dynamics codes. A large-scale modular code system with many options called ARC has been under development at ANL for some years. Another modular system, JOSHUA, is under development at the Savannah River Laboratory.

The U. S. efforts in compilation and evaluation of criticality data for fast reactors are still in their infancy. The chief data sources are ANL plate criticals,* Los Alamos homogeneous criticals,[93] and data from critical experiments performed abroad. Not all these data are readily accessible, and there are many gaps and inconsistencies in the available literature. In 1968 BNL undertook the compilation of fast criticality data.[94] A three-volume compilation of available data from ZPR-3, ZPR-6, and ZPR-9 experiments has been completed. In view of widespread interest, it is hoped to publish this work, although funds for its continuance have been cut off.

The evaluation of fast reactor integral data is also in a preliminary state as evidenced by many recent papers, e.g., at the 1970 Annual Meeting of the American Nuclear Society.[95] One reason is historical. The first ANL fast breeders studied had hard spectra

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*Information concerning these experiments is contained in ANL Progress Reports. A recent ANL report series (ZPR-TM-) contains data on a number of ZPR criticals and future work schedules. See C. E. Till and W. G. Davey, The Demonstration Benchmark Critical Assemblies Program, *Transactions of the American Nuclear Society*, 13(1): 293 (1970). A "rod core" has also been built to compare heterogeneity effects between plate mockups and actual rod configurations.

where heterogeneity effects were not serious and Doppler coefficients were very small. On the other hand, fast power reactors will be large, dilute systems where heterogeneity effects are important. Heterogeneity effects are even more complex in large-volume critical experiments with the usual lattices composed of drawers containing a variety of platelets.

Consequently efforts to improve the methods used to analyze experiments in these lattices are under way. Current methods of treating resonance self-shielding and sodium-void[96] effects are proving inadequate to analyze criticals such as ZPR-6 Assembly 5, a 2700-liter UC core. The analysis of the material worth (danger coefficient) of samples placed in the center of an assembly is also turning out to be quite complex.[97] The analysis of Zebra experiments at Winfrith has shown that the influence of the sample on the flux in the surrounding medium has to be taken into account (M. M. Levine, personal communication). The material-worth integral experiment is important because of its precision and because it could shed light on the adequacy of cross-section data sets for many materials.

The fast critical ZPR-3 Assembly 48 is one of the "benchmark" experiments being studied by the CSEWG committee. First objectives were to test ENDF/B Version I data, and to compare cross-section processing codes and diffusion-theory and transport-theory codes in use at various laboratories. The initial results showed considerable variation. Some of the differences were readily discovered, such as an erroneous treatment of $(n,2n)$ events in the ANL–LASL MACH-1 diffusion-theory code and differences in various versions of the MC^2 cross-section processing code.[98] As suggested earlier, methods of group averaging in the resolved resonance range are sensitive to the weighting functions used and their accuracy needs further investigation. Differences between MC^2 and other processing codes such as GRISM are likewise being traced.[99]

The MC^2 code itself has a number of weaknesses at present. Owing to limitations on P_n scattering ($n = 0,1$ only), the code is inadequate for the analysis of hard-spectrum criticals.[85] The so-called narrow resonance (NR) approximation[19] for resonance absorption used in MC^2 is inadequate not only for low-energy resonances in ^{238}U but also, as has been known for some time,[100] for the important 2.85-keV resonance in sodium. A code to replace MC^2, alleviating these problems, is in preparation at ANL.

Heterogeneity experiments in a fast reactor spectrum have been performed at BNL with various foils to determine the approach to an equilibrium spectrum in a lattice cell containing enriched-uranium, stainless-steel, and graphite plates. The results showed the importance of determining deviations from equilibrium. The importance of determining the moisture content of graphite in fast critical experiments was also shown.[101] In plutonium-fueled critical experiments with substantial ^{241}Pu content, the age of the fuel may also be significant since ^{241}Pu has a lifetime of only 14 years.

REACTOR KINETICS

Reactor kinetics is a large subject, so we restrict ourselves to a brief history, a statement of the major problems, and references to important sources and review papers.

In the past, reactor-kinetics studies have followed major problems uncovered during reactor design or operation. The importance of delayed neutrons to stability of reactor operation was realized from the start, and considerable data have accrued on the yields, lifetimes, and energies of the delayed-neutron emitters. Known results to 1965 are reviewed in a book by Keepin.[102] Work in this area is continuing because of the importance of delayed neutrons in power reactors and their potential importance in the nuclear materials safeguards program.

The importance of the Doppler coefficient as an inherent shutdown mechanism was also realized at an early date. The first deliberate set of reactor excursions[103] controlled solely by the Doppler coefficient was carried out at BNL in 1950. Simpler means of measuring Doppler coefficients have since been developed at ANL, AI, and European laboratories,* but the deliberate excursion experiments are still very useful, although they were generally carried out at remote sites. These sites include Godiva, KEWB, BORAX, SPERT, and TREAT.[104] The last two facilities continue to yield physics and materials information important to reactor safety, including rough limits on energy inputs above which damage to fuel elements results.[105] Work at the SEFOR reactor, which has just completed its low-power tests,[106] includes excursion experiments. SEFOR fuel was designed to facilitate the determination of its Doppler coefficient at temperatures near fuel melting.[107]

Interest in reactor safety was stimulated by a series of early criticality accidents. Such serious incidents as those at NRX in Chalk River, Windscale in England,

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*See Ref., 20, Vol. II, pp. 343–370.

and SL-1 at the National Reactor Testing Station in Idaho led to greater research efforts to understand and protect such reactors. Not the least of the knowledge gained was experience in coping with the aftermath of these incidents. The EBR-I meltdown had its beneficial aspects since it exposed the danger of relying on delayed negative-temperature coefficients[108] and demonstrated the importance of preventing fuel-element bowing that leads to positive reactivities.[109] Since Bethe's work[108] laid considerable stress on the importance of determining reactor transfer functions, much research was encouraged on oscillator tests, noise analysis, stability analysis, etc., which has since provided considerable expertise in these areas.

Fast reactor safety is regarded as so important that one major manufacturer has suggested the use of BeO in prototype breeders to soften their spectrum and thus increase their Doppler coefficient. Coupled fast thermal systems and "spoiled geometry" breeders have also been recommended for their improved safety characteristics. Many large conferences on fast reactors include papers related to reactor safety.[110-112] The classical paper on energy release in a fast reactor core collapse is that of Bethe and Tait.[113] The model has since been greatly improved, and more local modes of failure have been examined. Much of the basic knowledge needed for rigorous calculations is not available, so that reliance is placed on empirical models and engineering tests.

The calculation of long-range reactivity effects, sometimes called burnup or depletion calculations, has kinetic aspects. By the end of the Manhattan Project, enough was known about xenon, samarium, and gross fission-product poisoning for rough depletion studies. As will be discussed later, the accuracy of these calculations is very important to the nuclear power industry.

Xenon override problems became severe as high-flux research and test reactors came into being. Xenon instability is important to modern thermal-power reactors and will be discussed later.

Control-rod theory,[2] later further developed by Hurwitz and others, is no longer generally regarded as intrinsically different from fuel-rod theory since modern computational tools apply equally well to both. Protection against conceivable accidents, such as rupture of a primary coolant pipe or failure of a control-rod mechanism, involves typical transients considered in power-reactor design. The results are used to determine required control-rod patterns and insertion rates. In addition to control rods, the modern PWR uses soluble boron to provide more uniform control

and may use burnable poison and part-length rods (for power shaping and axial xenon control).

Control in other reactor types may differ widely. In the water-boiler reactor, radiolytic gas formation is an important, inherent shutdown mechanism; in TRIGA and SNAP reactors it is the zirconium hydride moderator; and in fluid-fuel breeders it is fuel expulsion. In Brookhaven's HFBR (High Flux Beam Reactor), reliance is placed on reflector control, and there are no control rods in the core itself.[114] Greater detail on control problems can be found in standard texts.[115,116] Decay-heat transients are important in high-power reactors under normal shutdowns and under accident conditions. Way and Wigner[117] made the first study of the time dependence of radiation from fission products.

Early work on feedback effects during transients utilized quite simple models, such as that of a prompt negative-temperature coefficient with no heat loss (Fuchs–Nordheim model), with constant power extraction, or with Newton's law of cooling. One-velocity equations were used, and the role of the delayed neutrons was simplified. Much of this work is reviewed in the early Geneva conferences.[118,119] Studies of nonlinear effects were also carried out, again with simple physical models, and were based on "point"-reactor ordinary differential or integro-differential equations or "line"-reactor partial differential equations, again in one-energy group.[120] The line-reactor equations were used to study such questions as the distribution of delayed-neutron emitters in circulating-fuel reactors or transient effects of a 1D wave nature, such as a sudden inlet coolant-temperature change. The line was generally an open loop. The line model is still used in industrial studies of water-reactor dynamics and control, but the "line" becomes a closed loop including subloops, which can extend as far as the turbine in studies of pressure control of BWRs.[121]

The point-reactor model is still in wide use for transient studies by industry. Westinghouse uses a code called CHIC-KIN. It incorporates all known major feedback mechanisms and has been checked against SPERT transients.[122] General Electric has also described its methods of analysis for prompt critical excursions.[123] The codes used vary from very simple ones to an early version of WIGLE.

There exist a number of 1D diffusion-theory kinetics codes, including WIGL-3,[124] GAKIN,[125] RAUMZEIT,[126] and NOAH[127] for thermal reactors and QX-1[128] and FORE-II[129] for fast reactors. The WIGLE codes have had the most general use, but they

are slow and permit only a crude treatment of feedback effects. TWIGL is a similar 2D code.[130] There also exists a 1D, 2-group transport-theory code,[131] and a time-dependent version of ANISN is nearing completion.

Much effort is going into studies of methods of programming kinetics codes to reduce costs or to increase accuracy. In addition to more efficient numerical methods, these codes include novel nodal (mesh) methods or modal methods in which the flux in each energy group is typically approximated as a linear combination of products of separable time and space functions. A number of reviews of these methods are available.[132-134]

One expects that the principal tools in the field will remain based on few-group diffusion theory and mesh methods. The methods will include point-reactor calculations, with improved techniques for recalculation of the flux shape and spectrum at appropriate time steps, line-reactor calculations for single-channel studies, and 3D codes with a limited number of mesh points where required, as in studies of the possible propagation of fuel-pin failures in fast reactors or xenon control in thermal reactors. More sophisticated codes involving many energy groups, neutron-transport effects, or detailed lattice transients will be used, if they are cheap and flexible, to improve results from the more routine codes. This has been the experience in reactor-statics calculations and is emerging as the pattern in water-reactor kinetics.

In contrast to the intensive effort on code development, there are few studies of the basic physics questions that underlie assumed feedback models and other important reactor problems. These include such questions as void distributions in transient two-phase flow, component vibrations from fuel elements to heat-exchanger tubes, fuel-failure mechanisms, and radiation-induced swelling of stainless-steel cladding. The basic questions lie in the realms of hydrodynamics, continuum mechanics, and the physics of the defect solid state. Work is indeed being done on these problems but primarily from an engineering viewpoint.

Work on neutron dynamics and control and on neutron noise, waves, and pulse propagation has been reviewed at a large number of topical meetings. A book of review articles on many important areas related to reactor safety is also available.[135]

The AEC supports a major program on Water-Reactor Safety.[136] A summary of recent work has been published.[137] At BNL a Technical Assistance group for the AEC regulatory divisions investigates reactor-physics questions related to the safety of specific power reactors and thus helps speed up these reactors.

INDUSTRIAL PRACTICE

The day of reactor-design calculations by handbook and slide rule has long since passed in the industry. Every major manufacturer has a package of codes that is flexible enough to treat everything from complete plant lifetime studies and safety, control, burnup, and fuel-management studies to fuel-inventory and plant-cost analyses. The adequacy as well as the costs of their code packages is of great financial importance to the manufacturer. For the smaller companies who cannot afford this expense, there exist computer-service organizations that provide expertise with comparable code packages.

Most of the codes used by the various manufacturers are in the public domain, but some refinements in code development, cross-section modification, thermal—hydraulic recipes, control, fuel management, or operating experience may be regarded as proprietary. The volume of restricted data may be more than is necessary or good for the field as a whole. In Britain the various consortia assembled to build Magnox reactors pooled their common reactor-physics problems for solution in a cooperative effort with the United Kingdom Atomic Energy Authority. This decision apparently has not affected their competitive position in bids or tenders for current nuclear power-plant design and construction.

Descriptions of the codes and methods used by manufacturers as well as considerable additional design information can be found in their PSARs (preliminary safety-analysis reports) for specific power plants. Thus the PSAR for a modern BWR[138] states that a three-energy-group diffusion-theory model is used for gross reactor calculations. Multigroup cross sections are calculated by an approach similar to that in the MUFT programs.[139,140] Homogenized cross sections are obtained for fuel assemblies or groups of fuel assemblies, depending on the coarseness required in the diffusion-theory mesh. The homogeneous thermal-spectrum calculations use the heavy-gas kernel for water and energy-dependent transport cross sections.[141] Epithermal cross sections are matched to measured resonance integrals and age data. Burnup calculations are carried out by integrating the time-dependent fuel-depletion process under the changes in neutron spectrum typical of operating conditions. Gross core-power distributions and reactivity effects are generally calculated in 1D and 2D geometries with discrete and

homogenized control-rod representations. In addition, 3D power distributions, using a coarse-mesh, one-group representation are utilized.[142] Doppler, moderator temperature, and void reactivity coefficients are obtained from empirical recipes. The fact that spatial xenon oscillations are well damped by negative power coefficients because of the strong feedback effects of void formation in current BWRs is stressed. The verification of these standard methods and data against operating-reactor and critical experiments provides the precision necessary for reactor design.

The information contained in PSARs may not be particularly up to date. Large sections may be lifted bodily from older PSARs. Greater detail and information on advances in methods and codes is provided in occasional publications. Recent work at General Electric on the development of 3D capability for statics calculations and progress on codes for the calculation of xenon transients has been reviewed by Crowther et al.[143,144] Other transients important to BWR operation and safety have also been discussed.[145]

One of the problems discussed in Ref. 145 is that of obtaining a minimum value for the maximum power peaking throughout a fuel cycle by attempting to maintain a stationary power distribution. The stationary power shape (the so-called Haling shape[146]) is uniquely determined for any assumed control-rod pattern at the end of the cycle. Attempts to maintain low peaking factors in the Dresden 1 reactor and in the Garigliano (SENN) Station[145,147] were not successful. Although thermal margins in the latter plant were quite large, economics would dictate smaller margins as long as safety requirements are not compromised. There is, therefore, considerable incentive to increase the precision of present design methods to permit reactor operation closer to optimal conditions on the basis of such criteria as power peaking factors and fuel burnup.[148,149]

Industrial practice in PWR reactor-physics calculations varies to some extent since a larger number of vendors are involved. For details on the present state of water-reactor calculations at Westinghouse, the reader is referred to the final safety-analysis report (FSAR)[150] for Indian Point Unit 2, particularly Secs. 3.2 on reactor design and 14.2.6 on the current state of the art of transient analysis. The basic tool in reactivity, power distribution, and depletion analyses is the LEOPARD-PDQ-7 (or the earlier AIM-5) sequence, with proprietary refinements related to user convenience, higher isotopes representation, and temperature feedback. For a very detailed analysis, a full 3D, few-group, nuclear—thermal—hydraulic program,

THUNDER,[151] is available; it is clear that this code is not used for routine calculations. Experience on the IBM-7094 indicates that a 10,000-mesh-point problem can be solved in 2 hr at a cost of about $1000, and it is estimated that the same cost would permit the use of 30,000 mesh points on the CDC-6600.

THUNDER indicates the trend in industry toward greater detail in the spatial representation of large, complex cores, such as those being proposed for water reactors in the 1000-MW(e) range. Greater detail in neutron energy and angular distributions would only increase the cost, as would greater detail in transient studies. It remains to be seen what impact the next generation of such large U. S. computers as the CDC-7600 and the IBM 360-195 will have on these costs.

In the larger PWRs now under construction, axial xenon oscillations would occur in the absence of control action, and other oscillations are marginally possible. Hence part-length control rods have been introduced to suppress incipient oscillations. We do not intend to discuss this problem at any length since a review article by Weston M. Stacey, Jr., entitled "Xenon-Induced Spatial Power Oscillations" appeared in *Reactor Technology*, Vol. 13, No. 3, pages 252 to 279, and an article[152] on Savannah River experience is in press. Recent calculations of xenon-induced spatial oscillations in large PWR cores have been carried out by Westinghouse.[153] The results of a 3D calculation for a homogenized core (due to limitations in the code and running-time costs) were compared with 1D calculations for the axial oscillation and 2D calculations for the $X-Y$ oscillation in an average enthalpy plane. Although the stability predictions of the simpler models were consistent with the 3D calculation, there was considerable difference in calculations of the damping rate for the $X-Y$ oscillation and the antidamping rate of the axial oscillation. Clearly, the problem of xenon control, under the manifold restraints of operating a large PWR, will be around for some time.

At Combustion Engineering the basic lattice code is CEPAK, which is made up of MUFT, THERMOS, and a group of subsidiary operator codes that connect these two basic blocks and introduce approximations for resonance shielding, fuel-temperature effects, and improvements in the cell approximation. In addition, the CEPAK code performs fuel-depletion calculations using few-group cross sections with occasional respectralization in the multigroup calculations at given points in fuel life. Resonance capture is based on formulas incorporating a 0.7-b reduction in Hellstrand's correla-

tion for the resonance integral of ^{238}U and Levine's form of the equivalence principle[19] for lattices. Account is taken of the effect of fuel cladding and overlap of ^{235}U and ^{238}U resonances. Plutonium-240 self-shielding and Doppler broadening are based on analytical fits to ZUT[21] calculations. Since none of the fuel assemblies is entirely uniform, considerable work is continuing on the proper treatment of fuel-assembly boundaries, water-hole peaking effects, and non-uniform Dancoff corrections.

The flux mismatch that existed between MUFT and the standard version of THERMOS has been corrected in an approximate manner.[154] As mentioned earlier, the rigorous method of making this correction is to raise the upper energy cutoff in THERMOS as well as the accuracy of scattering kernels in the associated FLANGE system. Hellens (private communication) finds that correction of the flux mismatch now provides quite good agreement of calculations with Yankee–Rowe ^{240}Pu measurements. He states that the adequacy of the lattice code is very important since it is the basic source of information on fuel input and discharge inventories. As far as inventories are concerned, details of 2D and 3D spatial calculations appear to be of secondary importance.

For spatial calculations, Combustion Engineering uses PDQ-7 on its CDC-6600 computer as the basic tool because of its reliability, speed, and flexibility. The cross sections required for these diffusion calculations come from CEPAK, as do changes in cross sections with fuel depletion which are represented in the HARMONY adjunct to PDQ-7. For reactivity-lifetime calculations a rather coarse mesh is generally adequate and is used in analyzing operating reactors. The use of a coarse mesh requires awkward fuel-assembly heterogeneities to be incorporated somehow into CEPAK calculations, and these studies are continuing.

The normal design calculations employ a fine-mesh 2D calculation with PDQ-7 to evaluate thermal conditions in various fuel assemblies. Thus far 3D synthesis methods have not been used, since it is hoped that Combustion Engineering reactors can be operated with soluble boron control throughout core life so that the resulting power distribution will be nearly separable. Little difference has been found between coarse-mesh, 3D PDQ-7 calculations and those obtained by combining 1D and 2D depletion calculations.

In space–time calculations considerable work is being done on short time transients typical of safety questions. Some work is being done in the area of automatic control of possible xenon instabilities in

both the axial and azimuthal modes. For this purpose a high-speed, coarse-mesh, 3D program called NODCOD has been developed. NODCOD incorporates automatic control-rod motion for criticality, automatic control-rod insertion and removal for power shaping, and moderator- and fuel-temperature feedback. The code is still in the developmental stage, but it probably will play an increasingly important role in studies of automatic-control strategies. This will be true particularly if temperature coefficients are small at the high power levels now being considered. Reasons for concern about temperature-feedback effects are, of course, the reduction of thermal margins in the fuel to improve the economics of power production and the increased use of soluble boron which reduces moderator coefficients since some of the soluble poison is expelled when the temperature of the water is increased. Temperature-feedback effects are the only inherent restraints on xenon instability in large, high-flux PWRs.

The current methods employed by Babcock & Wilcox are described in the FSAR for the Oconee Nuclear Station.[155] Their standard depletion calculations are made with a 1D depletion package code called LIFE, which is a composite of conventional codes, MUFT-5, KATE-1, RIP, WANDA-5, and a depletion routine. The 1D WANDA calculation uses four-energy-group cross sections, which are obtained by collapsing a 34-energy-group MUFT calculation and the use of the Wigner–Wilkins heavy-gas model for the thermal constants. Disadvantage factors for the thermal group are calculated with THERMOS.

Similarly 2D calculations are done with conventional PDQ-5, PDQ-6, or TURBO codes. These calculations are carried out to determine the effects of strong localized absorbers such as their movable control-rod assemblies. Additional controls in Oconee include soluble boron, part-length axial-power-shaping (and xenon-control) rods, and, in Oconee 2, burnable poison rods. Burnable poison rods are used in Oconee 2 to compensate for fuel burnup and fission-product buildup to reduce total control requirements. The required properties of the 1D system are then matched to the 2D analysis. In this manner it is possible to analyze the simpler 1D system in a depletion survey problem with, it is stated, only a small loss in accuracy.

Current work at Babcock & Wilcox Co. involves the upgrading of their basic code library that apparently now includes PDQ-7 and its HARMONY adjunct. Work on FARED[91] has already been mentioned as has the experimental Physics Verification Program,[81] which is jointly supported by Babcock & Wilcox and the AEC.

An interesting aspect of the latter program is that it has yielded information on the response of solid, in-core detectors at temperatures approaching those of an operating power reactor.[156] There has been a long-felt need for in-core detectors for PWRs to supplement information obtained from out-of-core detectors and thermocouples since such detectors could add greatly to reactor safety. They will certainly be needed if automatic control of power reactors is to become a reality.

Recent progress has included the development of such so-called self-powered detectors as rhodium, which becomes an intense beta emitter upon neutron capture. With the increasing need for sophistication in the control and operation of water reactors, there is also an increasing need for greater knowledge of what is occurring at various points within the reactor core, which is virtually a black box. Fuel temperatures, for example, are not measured, nor are they readily calculable, since the degree and manner of cracking of ceramic fuel during operation, the gap width between fuel and cladding, and the concentration of fission gases within the gap cannot be stated with certainty. As a result, effective temperature- and power-coefficient recipes, based on operating experience, continue to be used in PWRs without adequate knowledge of their degree of conservatism. The logic behind a published Westinghouse recipe for power coefficients has been challenged.[157]

All water-reactor manufacturers relate their basic lattice codes and cross-section data to critical experiments and other integral data. Agreement with criticality data on enriched-uranium-fueled assemblies is generally quoted to within ±0.5% in reactivity. This agreement offers some reassurance in reactor design, although it may be based in part on a cancellation of errors of opposite sign. There would be greater reassurance if these results could be compared with unadjusted cross sections and rigorous calculations. However, the standard U. S. cross-section library ENDF/B still has a number of important uncertainties and omissions, as discussed in the body of this article, and thermal-reactor data have a low AEC priority. Rigorous calculations are also expensive, infrequently attempted, and then may come to grief because of an overeagerness to draw conclusions on the basis of an incomplete or insufficiently broad study. Concerning plutonium-fueled water reactors, there seems to be general agreement that the available data are rather sparse. Many of the data available have been obtained on small, high-buckling assemblies, which, as we have noted, require elaborate analysis. There is disagreement between industrial evaluators on the general adequacy of ENDF/B plutonium-isotope cross sections and concern about the adequacy of reactivity calculations for high burnup of ^{235}U-enriched or plutonium-enriched fuels.

For one to understand the approximations used in industry's basic code packages, it is instructive to review the work of Strawbridge and Barry[5] which underlies the popular LEOPARD code. The method is based on MUFT–SOFOCATE for the homogeneous calculations. A 54-group MUFT calculation in the B_1 approximation is used to cover the energy range from 10 MeV to 0.625 eV. The B_1 approximation is rigorous only to second moments in the slowing-down distribution and may therefore be inaccurate for high-leakage systems. The effects of neutron-migration anisotropy may also be significant.[158] The SOFOCATE code calculates the thermal-group constants averaged over a Wigner–Wilkins (heavy gas) spectrum below 0.625 eV. Modern improvements over the heavy-gas model for water reactors include the Nelkin and Haywood models. However, thermal-group cross sections are generally not too sensitive to thermalization models, provided integral quantities are preserved. Thermal-disadvantage factors are obtained by modification of a formula due to Amouyal and Benoist. As previously discussed, the THERMOS–FLANGE sequence is gradually replacing such recipes in industrial use. To pick at a fine point, we know that even THERMOS becomes somewhat inaccurate for very tight lattices.

In the resonance range, self-shielding factors are neglected except for ^{238}U. The neglect of self shielding for ^{235}U is explained on the basis that it applies to both capture and fission and hence has a small effect on the multiplication factor. These effects are, of course, of greater importance in fuel-burnup calculations. The lattice self-shielding of ^{238}U is calculated by a recipe that is fitted to Hellstrand's correlation. At higher energies, fast-advantage factors (i.e., ratios of fuel flux to moderator flux) are computed over the energy range from 10 MeV to 0.821 MeV only. Such factors are still important at lower energies. A comparison of the recipe used with results obtained by Rief's MOCA code indicated a considerable underestimate of the fast-advantage factor. In part, this discrepancy may be due to the choice of cross sections, particularly inelastic cross sections for ^{238}U. It is ironic that this old problem[11] still persists for lack of sufficiently accurate data.

The Strawbridge and Barry criticality calculations were compared with experimental data on a large number of UO_2 and uranium–metal–water lattices.

The average multiplication factor was 0.996 ± 0.007 for the uranium-metal cases and was 0.990 ± 0.009 for the UO_2 cases. The uncertainties in some of the reported experimental bucklings were noted without any attempt at their improvement. Later work has shown how to improve the analysis of leaky assemblies.[83,84]

Recent work[74,75] on the analysis of uranium–water lattices has placed greater reliance on Monte Carlo calculations. In addition to buckling data, reaction-rate measurements were analyzed. However, results were reported on only four metal-rod, aluminum-clad lattices and three natural-uranium slab lattices. The differences between Bettis data and ENDF/B cross sections[73] for ^{235}U below 0.625 eV were noted but not explored in detail. The methods of analysis used are a considerable improvement over the typical industrial survey that is used to adjust cross-section libraries. However, some of our reservations concerning the rigor of these analyses apply here also, particularly for the tight-slab lattices where leakage corrections are very large.

In summary, reactor-physics methods for power reactors have become more detailed as the reactors have become more complex; there is now an almost complete dependence on fast computers, codes, tapes, and disks. Much demand exists for experienced programmers and applied mathematicians. Unfortunately this trend has been accompanied by a decreased demand for the creative physicist and innovative engineer. In the first two decades after World War II, the latter made possible the great advances in reactor technology, in the use of new and exotic materials, in instrumentation and the growth of the scientific computer, and in the design and construction of novel reactors for research, testing, power, and propulsion. Today the national program in reactor technology is rapidly shrinking into a single, centrally directed project. This is not a natural evolution.

Future U. S. energy needs should have a high priority, and nuclear power is the only proved, long-term source for these needs. In particular, the breeder reactor will be built, even if announced goals on fuel burnup and costs are not completely met. The only possible future competition would be cheap, excess neutrons from D–T reactions in a controlled, thermonuclear reactor or spallation neutrons from a competitive, high-energy particle accelerator; such developments, however, are by no means certain, and if they do evolve, would be used to complement rather than displace the breeder.

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Solidification and Disposal of High-Level Radioactive Wastes in the United States

By K. J. Schneider*

THE SOURCE OF THE PROBLEM

In nuclear fuel cycles the irradiated fuel is discharged from the reactor with enough fissile or other valuable materials remaining to warrant their recovery for reuse. These spent fuels are aged for a few months to a year to permit decay of the short-lived radionuclides. The fuels are then shipped to a fuel-reprocessing plant where the valuable fissile and fertile fuel materials are separated from the fission products. The recovered fuel materials are returned to the fuel cycle for reuse.

In current fuel-reprocessing schemes, the volatile or gaseous radionuclides present in the spent fuel (e.g., ^{129}I, ^{131}I, ^{85}Kr, ^{131m}Xe, ^{133}Xe) are volatilized, and the resultant gaseous stream is treated for removal of some of these nuclides to meet emission limits. (Tritium is in this general category of volatile radionuclides, but it generally follows the water stream in reprocessing plants.) Approximately 99.9% of the remaining fission products are collected in the waste from the first product-decontamination step as a nitrate-based high-level aqueous waste. This high-level aqueous stream is concentrated, then stored in large underground tanks, either in the acidic form or after being made alkaline with caustic.

The wastes must be stored for centuries to permit decay of the major long-lived radioactive fission products (primarily ^{137}Cs and ^{90}Sr) to nontoxic levels; the small amount of plutonium present as a waste loss (usually about 0.1 to 0.5% of that in the original fuel) and some long-lived fission-product radionuclides require thousands of centuries to decay to nontoxic levels.

Obviously the control of these wastes to prevent them from entering man's biosphere in significant quantities for the long time periods involved is a formidable task. Currently the only practical means to prevent the radioactivity from entering our environment is to store the material under carefully controlled conditions until the radioactivity has decayed to nontoxic levels. Methods that may be better than storage might some day be developed. At this time, however, major technological advances are required for development of new schemes.

Interim storage of these high-level wastes as liquids has been done safely in the young atomic power industry. However, as the industry expands, storage problems will become increasingly complex. Consequently further improvements in the safety of waste storage in the future have been sought on a worldwide basis. Most schemes developed for this goal include conversion of the aqueous wastes to solids, followed by long-term storage in man-made vaults or disposal in deep geological formations such as salt. Although the relative safety of solids vs. liquids has not been quantified, it is generally recognized that storage as solids would be considerably safer. The increased safety of storage as solids is due to the decreased mobility, decreased leachability in water, smaller volume, and the greater physical ruggedness of encapsulated solids compared to liquids. This analysis has been emphasized by the U. S. Atomic Energy Commission (AEC) in its proposed policy in the *Federal Register*[1] for all U. S. commercial fuel-reprocessing plants, as follows:

1. A commercial fuel-reprocessing plant's inventory of high-level liquid radioactive wastes will be limited to the quantity produced in the prior 5 years. High-level

*Battelle–Northwest, Richland, Wash. 99352.

liquid radioactive wastes in excess of this authorized inventory must be converted to an AEC-approved solid form.

2. All high-level radioactive wastes must be transferred in the approved solid form to a federal repository as soon as practicable, but in no event later than 10 years following separation of fission products from the irradiated fuel. Federal repositories, which will be limited in number, will be designated by the AEC.

3. Upon receipt of the solidified wastes, the federal government will assume physical responsibility for these radioactive waste materials, although industry will pay the federal government a single fee designed to cover all costs of disposal and perpetual surveillance. Before retirement of the commercial reprocessing plant from operational status and before termination of licensing, transfer of all such waste to a federal repository must be completed.

4. Ultimate disposal of high-level radioactive fission-product waste material will not be permitted on any land other than that owned and controlled by the federal government.

The predicted quantities of high-level wastes[2] to be produced during the next generation are shown in Table 1. The amount of high-level waste materials handled by commercial enterprises in the year 2000 is expected to be about 1000-fold greater than that available today.

HIGH-LEVEL LIQUID WASTE

High-level radioactive wastes are those which contain at least 1 Ci/liter of liquid or 70 Ci/kg of solid.[3]

These values, which are used in this review, are high enough to require thick radiation shielding (equivalent to several feet of normal concrete) and to require cooling of the significant heat evolution from decay of radioactive fission products.

Chemical Characteristics

Figure 1 shows schematically the range of compositions of most chemical constituents that may be present in high-level wastes from light-water reactors (LWR) using variations of the Purex solvent extraction process.[4] The amount of material is shown as kilograms of oxide per metric ton of fuel that would result if the liquid waste is converted to a solid. All constituents except fission products and aluminum from alloy fuels are added during the reprocessing steps. The variations shown represent typical ranges arising from variations within the fuel-reprocessing scheme. A "clean" waste (i.e., where almost no salts are added during fuel reprocessing) and a moderately "dirty" waste are superimposed on the figure. With moderately clean wastes, the fission-product content in the waste can be much greater than that of the "inert" salts. Typically, the wastes are nitrates of sodium, iron, and fission products, with smaller contributions of other constituents, such as chromium and nickel. Sulfate can be present in significant quantities with some reprocessing schemes. The compositions shown in Fig. 1 are based on current reprocessing technology and on the assumption that the fuel cladding is not dissolved with the fuel; consequently the fuel-cladding constituents are not present in the high-level waste.

Table 1 Estimated Growth of Nuclear Power and High-Level Wastes*

	Calendar year			
	1970	1980	1990	2000
Installed electricity, 10^3 MW(e)	6	150	450	940
Fuel processed, 10^3 metric tons/year†	0.055	3.0	9.0	19
High-level liquid wastes, 10^6 gal/year‡	0.017	0.97	3.3	5.8
Accumulated high-level wastes				
As liquid, 10^6 gal‡	0.4	4.4	29	77
As solid, 10^6 gal§		0.33	2.1	5.8
Accumulated fission products				
Weight, metric ton	2	550	4,200	14,000
Beta activity, MCi	2,000	19,000	103,000	267,000
Heat-generation rate, MW	0.7	55	280	730
^{90}Sr, MCi	4	960	5,700	12,000
^{137}Cs, MCi	5	1,280	8,000	20,000

*From Ref. 2.
†Based on 33,000 MWd/metric ton.
‡At 100 gal/10,000 MWd(t).
§At 1 ft^3/10,000 MWd(t).
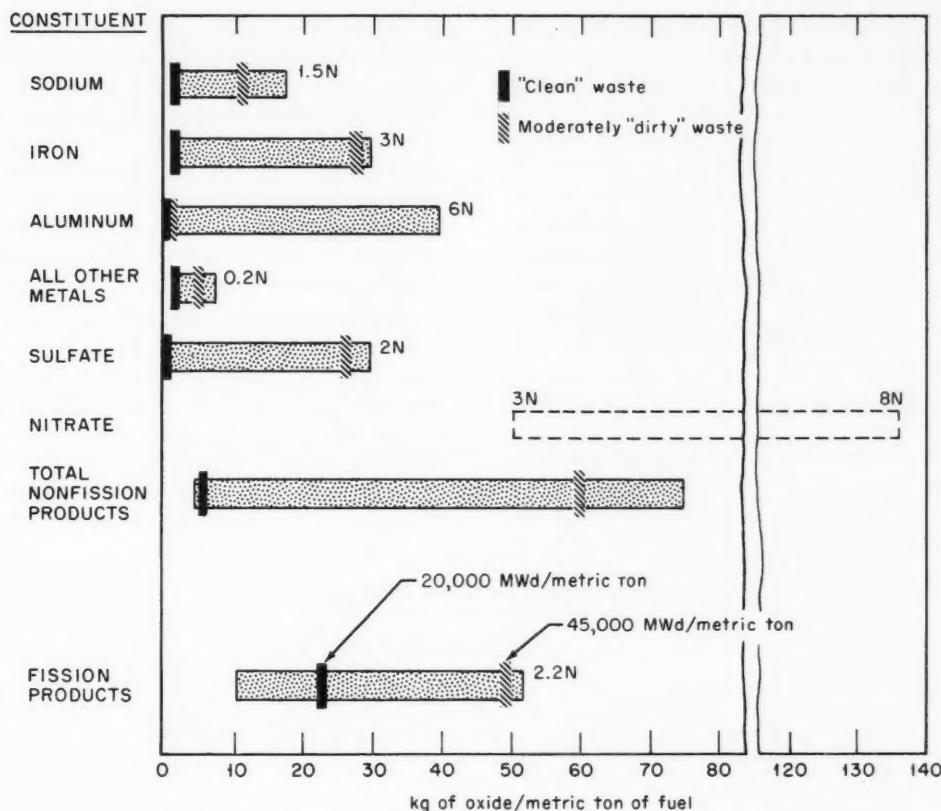
NOTES: NORMALITIES ARE FOR 100 GAL OF LIQUID WASTE/metric ton.
NITRATE IS NOT PRESENT AFTER WASTE IS SOLIDIFIED.

Fig. 1   Ranges of chemical compositions of high-level wastes.

Waste compositions from the reprocessing of fuel from liquid-metal fast breeder reactors (LMFBR) are expected to be similar to those shown in Fig. 1. The major potential difference is the larger concentration of fission products (and the resultant increased radio-activity levels) present in the core fuel. However, current plans generally favor reprocessing combined core–blanket fuels together, which results in fission-product concentrations generally within the range shown. An additional difference in the wastes from fast reactors is that the relative amounts of the potentially volatile fission products Ru, Se, Te, Sb, and Sn will increase (although the total amount of the latter four will still be relatively small).

Wastes from reprocessing of fuels from other types of reactors, such as high-temperature gas-cooled reactors, light-water breeder reactors, and molten-salt reactors, may have different compositions. Because the compositions, volumes, and timing of wastes from these reactors cannot be quantified at this time, they are not discussed here.

As indicated in Fig. 1, wastes can be either stored with as much as $6M$ free nitric acid, denitrated to as low as about $0M$ free nitric acid, or made alkaline with caustic, with or without prior denitration. Control of nitrate or acid concentrations may be maintained to control corrosion rates or oxidizing potential in waste processing or storage. Minimum waste volumes are based upon solubility limits of salts that might precipitate and cause handling problems. Total metallic-ion concentrations are generally in the range of 4 to $8N$.

Approximately 15% of the fission products present in high-level wastes after 6 months out of the reactor will eventually decay to other chemical elements, many

of which will have different chemical valences and chemistry than their precursors. Consequently chemical compositions of high-level wastes will be somewhat different after long-term storage than initially.

Another important point of interest is that the actual weight of fission products is significant, as indicated in Table 1. Approximately 0.9 kg of nongaseous fission products result from each 1000 MWd of thermal nuclear energy. In addition, transuranium and transplutonium elements (americium, curium, neptunium, etc.) and plutonium (as a waste loss) are present to the extent of approximately 0.04 kg/1000 MWd of thermal energy.

Fission-Product Content and Heat-Generation Rates

One of the most striking characteristics of high-level wastes is that they contain essentially all of the intensely radioactive, nonvolatile, fission products in the irradiated fuel. The primary fission products are a mixture of more than 30 elements with atomic numbers ranging from 30 to 64. Most of these fission products are radioactive, and more than 300 different isotopes have been observed.[5] In high-level wastes only the stable and relatively long-lived radioisotopes are significant chemically or radioactively. As shown in Table 2, the number of radioactively important nuclides reduces to less than a dozen, and the total number of chemically important nuclides reduces to about 20.

Fission products in Table 2 were calculated for a fuel exposure of 45,000 MWd/metric ton (at 30 MW/metric ton),[6] which is believed to be a probable maximum for future thermal reactors.[7] For comparison the last column in Table 2 gives values for a fuel exposure of 20,000 MWd/metric ton (at 15 MW/metric ton), which is typical of current discharges from power reactors. For additional comparison the amount of

Table 2　Major Fission Products in High-Level Wastes

| Element | Av. atomic weight | After 1 year* | | After 5 years | | After 1 year moles/liter (378 liters/metric ton) | |
		Ci/metric ton	g/metric ton	Ci/metric ton	g/metric ton	45,000 MWd/ metric ton	20,000 MWd/ metric ton*
Rb	86		463		470	0.014	0.007
Cs	135	3.68×10^5	3,952	19.09×10^4	3,677	0.077	0.037
Sr	89	1.05×10^5	1,191	9.16×10^4	1,124	0.035	0.016
Ba	138		2,184		2,459	0.042	0.017
Y	89	1.12×10^5	622	9.16×10^4	621	0.018	0.009
La	139		1,729		1,729	0.033	0.015
Ce	141	4.41×10^5	3,440	1.26×10^4	3,305	0.064	0.028
Pr	141	4.41×10^5	1,611	1.26×10^4	1,611	0.030	0.013
Nd	145		5,531		5,665	0.101	0.043
Pm	147	1.03×10^5	111	3.58×10^4	39	0.003	0.002
Sm	150		1,039		1,111	0.018	0.008
Eu	153	0.16×10^5	270		256	0.004	0.001
Gd	156		127		144	0.002	
Zr	93	0.26×10^5	5,050		5,116	0.143	0.064
Nb	95	0.55×10^5					
Mo	98		4,856		4,859	0.131	0.058
Tc	99		1,153		1,153	0.031	0.014
Ru	102	2.84×10^5	3,118	1.80×10^4	3,040	0.081	0.033
Rh	103		520		520	0.013	0.007
Pd	106		1,736		1,814	0.043	0.016
Ag	109		68		68	0.002	0.001
Cd	111		103		103	0.003	0.001
Sn	123		78		78	0.002	0.001
Sb	123		22		16		
Te	130		705		711	0.014	0.006
Total		19.5×10^5	39,679	45.3×10^4	39,689	0.904	0.397

*Exposure at a specific power of 15 MW/metric ton.

significant fission products in LMFBR core fuel[8] and the reference LWR fuel after 6 months' aging is shown in Table 3. Transplutonium elements present in the wastes in modest quantities are not shown in Tables 2 and 3. Additional information on fission-product and transplutonium elements has been published in Ref. 9.

The rate of heat generation in high-level wastes (arising from beta and gamma emission) is sufficiently high to require positive cooling. High-level aqueous wastes usually self-boil for many years in the absence

Table 3   Major Fission Products in Spent Fuels from LMFBR and LWR

| | Quantity, g/metric ton | |
Element	Fast reactor core fuel*†	Thermal reactor fuel‡
Se	166	68
Rb	609	462
Sr	1,310	1,201
Y	677	624
Zr	7,584	5,045
Nb	60	9
Mo	8,800	4,841
Tc	2,384	1,153
Ru	8,535	3,154
Rh	2,490	519
Pd	6,660	1,701
Ag	756	68
Cd	440	103
In	42	2
Sn	436	78
Sb	160	23
Te	1,666	704
Cs	10,830	4,003
Ba	3,540	2,134
La	3,058	1,729
Ce	7,110	3,518
Pr	3,125	1,610
Nd	10,168	5,453
Pm	1,015	127
Sm	2,470	1,023
Eu	407	273
Gd	323	127
Tb	30	2
Dy	20	
Total	~89,000	~40,000

*Does not include gases or elements present in quantities less than 10 g/metric ton in core fuel.

†For exposure of 100,000 MWd/metric ton at 200 MW/metric ton, 6 months of cooling.

‡For exposure of 45,000 MWd/metric ton at 30 MW/metric ton, 6 months of cooling.

of forced cooling. Aqueous wastes have been generated with radioactivity concentrations as high as 2500 Ci/liter. The decay heat associated with the radioactive fission products from fuel exposures of 20,000, 45,000, and 100,000 MWd/metric ton (for core fuels from fast reactors)[8] are shown in Fig. 2 as a function of time out-of-reactor. The rate of decrease of heat generation is substantial during the first 5 years of cooling when most of the moderately long-lived fission products have decayed to stable isotopes. The primary remaining radionuclides are the long-lived ^{137}Cs, ^{90}Sr, and some transplutonium isotopes.

Physical Characteristics

High-level aqueous wastes are mixtures of metallic nitrate salts and insoluble salts, or precipitates. In most high-level acidic wastes, the settled volume of precipitate ranges from about 3 to as much as 20% of the total volume. The precipitates are troublesome in that most waste solutions must be continuously agitated to prevent settling of the solids as layers of sludge with resultant plugging of equipment and formation of thermal hot spots.

Almost all acidic high-level wastes contain a precipitate of zirconium phosphomolybdate (the zirconium and molybdenum are fission products, and the phosphorus is from the solvent used during reprocessing). Fortunately this precipitate is amorphous and relatively free from caking.[10,11] If the nitrate concentration is high, insoluble nitrate salts of the fission products barium and strontium form precipitates. These solids are relatively unfriendly in that they are hard and crystalline.[12] If present, silicon will usually be in the form of fairly flocculent silicic acid.

The presence of sulfate in concentrations greater than about 0.5M causes precipitation of rare-earth sulfates of sodium. This precipitate is crystalline and tends to settle as a very rugged, cement-like cake that is extremely difficult to break up.[11] Sulfate also causes precipitation of relatively heavy sulfates of the fission products barium and strontium.

Most wastes contain a small amount of unknown precipitate (less than 1 vol.%) that is hard and crystalline. This small amount of unfriendly solid (which is known to contain some noble-metal fission products) contributes to excessive wear of pumps and other mechanical equipment.[13,14]

Wastes neutralized to the alkaline condition contain hydrated oxide precipitates of nearly all metallic constituents except the alkali metals. These precipitates are voluminous (initially 20 to 40 vol.% of the total liquid volume, settling to 5 to 10% after extended

Fig. 2   Heat content in spent nuclear fuel.

storage), hydrous, and flocculent, but contain sufficient amounts of crystalline solids to cause a high degree of caking if allowed to settle and age.

The presence of large amounts of phosphates in the wastes generally causes voluminous precipitates (up to 50 vol.%) that are normally relatively flocculent and easy to handle.[12]

High-level wastes have specific gravities ranging from 1.1 to 1.3, depending upon concentration and constituents. Viscosity is usually in the same range as water, and the boiling point varies from 103 to 115°C.

Experience with Storage

Aqueous high-level wastes have been stored in the United States for nearly 25 years. Reprocessing plants at three AEC sites are currently storing significant quantities of high-level wastes: Richland, Wash.; Savannah River, S. C.; and the National Reactor Testing Station (NRTS) near Idaho Falls, Idaho. Over 80 million gal of radioactive solutions and sludges, mostly in the alkaline condition, are stored in nearly 200 tanks at these three AEC sites.[15] Alkaline wastes are usually stored in mild-steel tanks; acidic wastes are stored in stainless-steel tanks. The only private source in the United States wherein significant quantities of high-level wastes are stored is the fuel-reprocessing plant near Buffalo, N. Y., owned and operated by Nuclear Fuel Services, Inc. (NFS). Approximately 600,000 gal of alkaline wastes are stored in one mild-steel tank, and 15,000 gal of acidic wastes are stored in a stainless-steel tank at the NFS plant site.[16]

These waste-storage tanks vary in capacity from 15,000 gal to the currently more common size of about 1,000,000 gal. The tanks are buried under about 10 ft of earth to take advantage of the earth cover as inexpensive shielding, as structural support, and as protection from surface weather conditions.

Figure 3 shows the main features of the latest design of a 1-million-gal tank by the Atlantic Richfield

storing acidic wastes (i.e., those at the NRTS site and some at the NFS site) contain internal cooling coils to maintain the waste temperature below about 65°C, above which corrosion can become significant.[18] Most tanks at Savannah River have a more shallow tank for the secondary container. The older tanks at Richland have no secondary tank. Tanks for nonboiling wastes whose levels of heat from radioactive decay have been
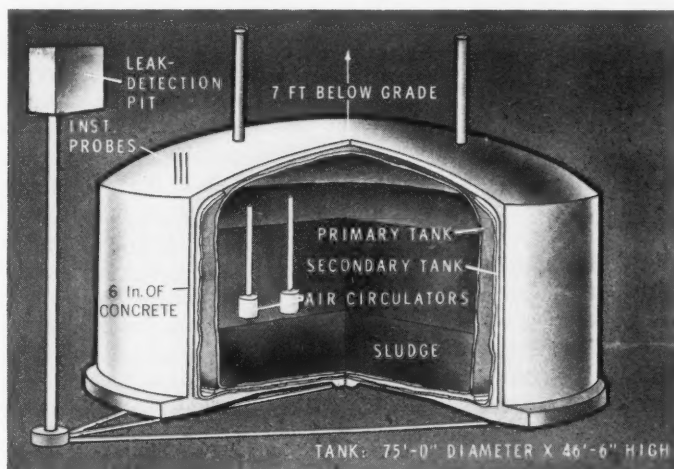
Fig. 3   New boiling waste tank.

Hanford Company at Richland, Wash. This design represents the latest in the evolution of improved features over the past 20 years. Here a large mild-steel tank is enclosed in a secondary mild-steel tank for additional safety. Both tanks are surrounded by a concrete structure. Up to 22 air-lift circulators are used to agitate the wastes to prevent settling of sludge with resultant formation of thermal hot spots. Lateral test wells, shown beneath the tank, permit detection of leaks into the ground. Not shown are the leak-detection system between the two tanks and the venting system wherein the gases and vapors from the self-boiling wastes are routed to external aerosol eliminators, condensers, and filters. The condensate is recycled to the storage tanks to prevent overconcentration and to aid in cooling by reevaporation. Alternatively, some of the condensate may be further treated to reduce radionuclide concentrations below disposal limits before being discharged to the ground.[17]

Some tanks at Savannah River and Idaho Falls contain water-cooled coils to keep the contents below boiling temperature. All stainless-steel tanks used for

significantly reduced by aging or by removal of heat-generating isotopes are generally similar except they have a simpler vent system and fewer ancillaries, such as instruments, monitoring systems, and recirculators.

Although incidents of loss of control of radionuclides have not occurred, a total of 15 tanks have failed in the United States.[15] All failures were in mild-steel tanks containing neutralized waste: 11 at Richland and 4 at Savannah River. No failures have occurred at the NRTS or at the NFS plant sites. All failures have occurred at stressed areas near welds, owing to stress-corrosion cracking accelerated by the nitrate—nitrite ions. Service life to failure has varied from 0.4 to 13 years, although most of the nearly 200 tanks (including many tanks more than 20 years old) are apparently in good condition. Leaking tanks are removed from service, and, unless the leak seals itself with a plug of high-density sludge, as frequently happens, the liquid level is dropped below the leak by transferring supernate to another tank. At both sites there is no evidence of radioactivity entering the

groundwater, and therefore no hazard to the public has resulted from the leaks that have occurred.[19]

Laboratory data indicate the mild-steel tanks (which are generally 0.25 to 0.875 in. thick, depending on location within the tank) should corrode at very low rates of about 0.00002 in./year if stress-corrosion cracking does not occur.[20,21] Past experience with mild-steel tanks has indicated an average useful life of 15 to 40 years. Recent application of in-place stress relieving of the completed tanks is expected to increase the life of the tanks.[19] Corrosion rates on 304L or 347 stainless-steel tanks by acid wastes are 0.0002 to 0.0005 in./year at $60°C$ but increase rapidly with increasing temperature to about 0.05 in./year at temperatures near boiling.[22-24]

DISPOSAL AND STORAGE

The ultimate objective in disposal of any type of hazardous wastes, nuclear or nonnuclear, is to permanently remove the materials from man's biosphere. Such removal is termed "ultimate disposal" in this article. Such disposal may be accomplished by one of two means: (1) convert the hazardous material to nonhazardous material, or (2) dispose of the material so it cannot enter man's environment.

For high-level radioactive wastes, potential solutions have been aimed at attainable schemes offering significantly improved safety by preventing the materials from entering our environment. All the potential schemes involve disposal or long-term storage of the wastes in a manner that will significantly reduce the possibility of the wastes' entering our biosphere for up to hundreds of thousands of years. The storage schemes incorporating prior conversion to solids are those which are generally considered the most credible.

Long-Term Disposal or Storage

Long-term disposal or storage (sometimes referred to as perpetual storage) is a feature common to all currently practicable methods for disposal. Although some of the storage schemes are recognized to be useful only for interim storage, they are included with proposed long-term storage schemes here. The primary methods in this category which have received attention are

- Disposal as solids in salt mines.
- Storage as solids in deep underground caverns.
- Storage as solids in man-made vaults.
- Disposal as solids in the ocean deeps.
- Disposal as liquids in deep wells.

- Perpetual storage as liquids in deep, underground caverns.
- Perpetual storage as liquids in tanks.

To many waste-disposal experts, the schemes considered to have the better potential for long-term safety are those wherein wastes are placed in certain deep geologic formations (500 or more feet deep) on land.[25-27] Placing the wastes in these locations should keep the wastes out of man's immediate environment for the long time required, the wastes would be less affected by future uncertain conditions on the surface caused by either man or nature, and reliance on human surveillance should be minimum. In addition, underland areas are generally more accessible than undersea areas, and much is known about the underland geology. To be likely candidates for disposal or storage areas, geologic formations must have been stable over the past million years or so, they should not indicate potential instability during the next million years, and they must offer one or more very reliable barriers between the wastes and the land, liquids, and gases in our environment.

One concern about any storage or disposal scheme is whether or not we have enough answers to be certain that stored wastes will remain "forever" out of our biosphere. Schemes offering retrievability permit undoing a problem that may not have been foreseen. For example, man may sufficiently alter the earth to the point of penetration of the essential barriers between the wastes and our environment. Also, truly ultimate disposal techniques may be developed to practicability within the reasonably near future, or man may someday develop a different and vital need for the storage area. On the other hand, retrievability in a storage system would normally require more surveillance, more control records, and more legislation to be enforced. Thus possible trade-offs of retrievability for other storage or disposal characteristics must be carefully weighed.

Disposal as Solids in Salt Mines. Disposal of high-level wastes as solids in salt mines has received more study in the United States than any other storage method. Extensive development and testing of this technique have been undertaken by workers at Oak Ridge National Laboratory (ORNL) since the National Academy of Sciences (NAS) committee recommended that disposal in salt was the most promising method for the near future.[25,26] This concept is expected to be put into use soon in the United States.

In this concept the waste is converted to an encapsulated solid, transported to the site of a previ-

ously excavated salt mine, and placed above the top of a shaft that extends down to the mine. The canned waste is transferred through the shaft to a shielded, manually operated carrier in the mine. The waste is then transferred to a room in the mine and placed in preexcavated vertical holes in the floor. Crushed salt is put into the hole to fill the annulus between the waste can and the wall of the hole and to fill the 6 to 8 ft of hole above the waste can for shielding. The process is continued until all the holes in a room are filled. Excess salt chips from the mining operation are then put back in the room. After a few decades the salt will plastic-flow under the pressure of the overburden to compress the crushed salt in the room to a monolithic formation, thus sealing the waste in salt. The concept is shown in Fig. 4 as developed by ORNL workers in Project Salt Vault, a radioactive demonstration in a salt mine near Lyons, Kans.[28,29]

The overall conclusions from the radioactive demonstrations of disposal in salt using spent fuel elements in place of radioactive waste during 1965 to 1967 were that the scheme was feasible, and sufficient data were obtained to make possible the design of a safe, long-term storage facility.[28] As a result, an ORNL study[15] is being made to determine the feasibility of establishing an operating salt-disposal facility in the mid-1970s.

The proposed use of salt disposal is based upon the premises that salt forms the final container for the waste (the original can is really needed only for transportation), salt is essentially impermeable owing to its plastic properties, salt formations are dry and contain no circulating groundwater, salt has no "communication" with the biosphere, and salt is a good conductor of heat. In addition, suitable salt formations are widely distributed and abundant, and the mining costs are relatively low. Bedded salt deposits are believed to be better than salt domes for waste storage.

Spacing of the waste containers (typically 5 to 15 ft apart) is conservatively based upon the heat generation rate in the waste to limit the maximum salt temperature to 200 to 250°C. At higher temperatures the small amount of water of hydration in the salt tends to break up the salt into small chunks at localized areas. The space requirement[30] for all the high-level wastes generated in the United States by the year 1995, aged 5 years to the year 2000 before storage, is approximately 1600 acres on the basis of the data in Table 1.

Mild-steel containers are expected to fail within a few years owing to corrosion from the salt, and the stainless-steel containers will fail in as short a time as a few months. From that point on, the salt bed becomes the container.

Estimated costs for disposal of solidified wastes in salt mines vary from about 0.003 mill/kW(e)-hr after 10 years of aging to about 0.01 mill/kW(e)-hr after 2 to 4 years of aging.[30]

Storage as Solids in Deep Underground Caverns. Storage of high-level wastes as solids in underground caverns (other than salt caverns) has received little study, although the report of the special NAS committee to provide an overview of potential storage or disposal schemes indicates that it is a potentially promising scheme.[25] In this case the waste would be first converted to solids and then packaged and transported to the storage site. There the waste would be placed into abandoned mines or specially prepared caverns 500 or more feet under the ground in hard rock such as granite or metamorphosed basalt. When a specific storage room became filled, it would be sealed by concrete and abandoned. The safety of this method would depend upon the stability and impermeability of the rock formation; the geology, hydrology, seismology, and climate of the surrounding area; the stability of the solid waste; and the integrity of the waste containers.

Preliminary cost estimates for this storage technique by workers at ORNL indicate storage costs would be about twice those of salt storage, or in the range of 0.01 to 0.02 mill/kW(e)-hr, primarily because of the higher excavation costs.[31]

Storage as Solids in Man-Made Vaults. Storage of solid wastes in man-made concrete vaults has been studied briefly by workers at Chalk River[32] and ORNL[31] and has been used at the NRTS since 1963. In this concept the high-level wastes are first converted to solids, then placed in large concrete rooms built under about 10 ft of soil to provide inexpensive shielding and to provide safety from the surface environment. Heat from the waste can be dissipated through the concrete, into the ground, and then to the surface, or it may be dissipated by forced- or natural-convection cooling using air. At the NRTS site[33] the granular solid waste is transported by air fluidization through pipes to the air-cooled storage tanks as shown in Fig. 5.

Storage in man-made vaults is generally considered to be useful only for interim storage since surveillance and controls are required. Storage costs for power-reactor wastes in concrete vaults have been estimated by ORNL workers to be about 5 to 7 times the cost of
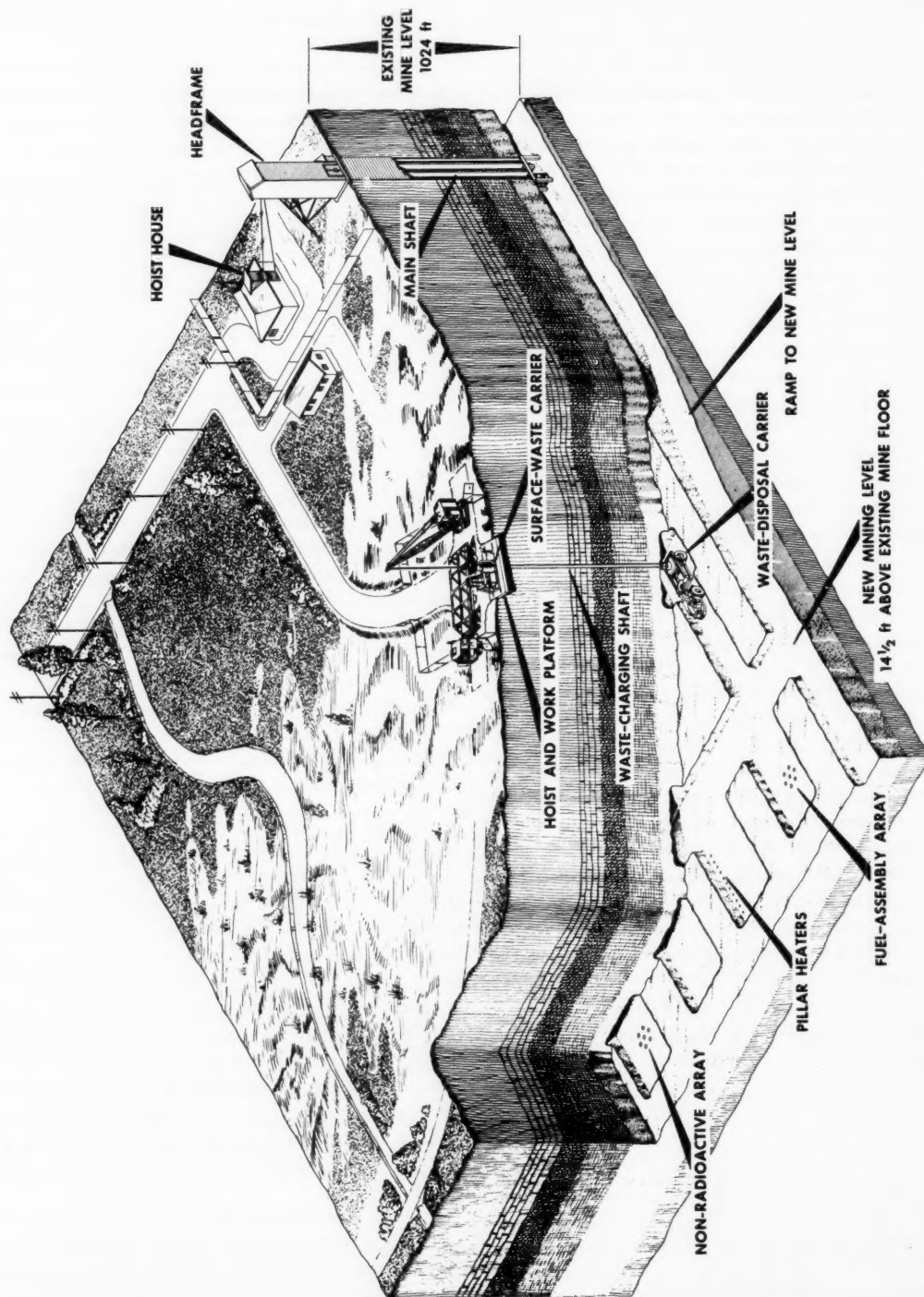
Fig. 4 Demonstration of radioactive solids disposal in salt.
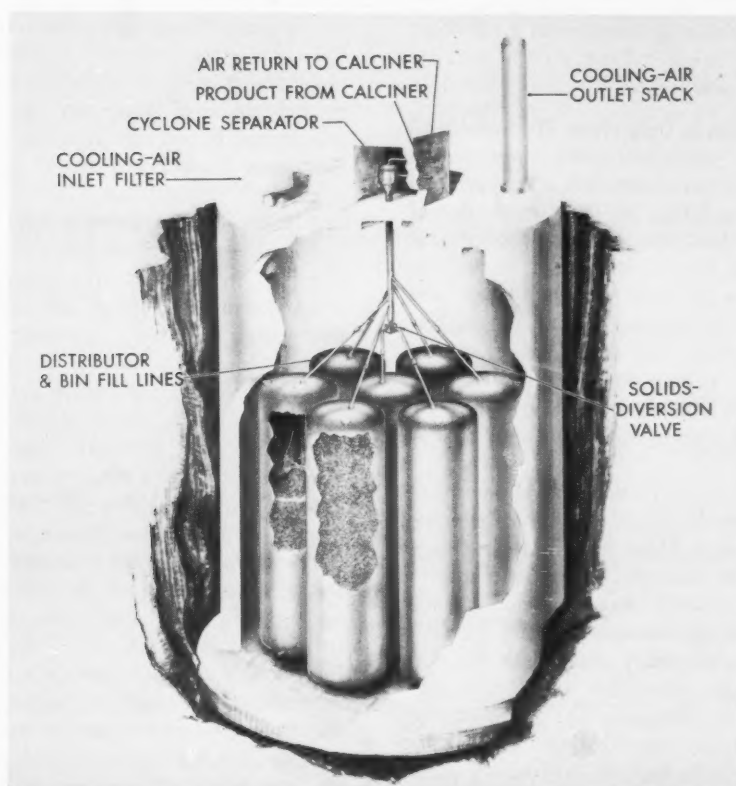
Fig. 5   WCF solids storage bins.

storage in salt mines, or in the range of 0.03 to 0.07 mill/kW(e)-hr, depending upon heat generation rates.[31] Total costs for storage of the granular solids at NRTS are $0.80 per gallon of original liquid waste[33] but cannot be compared directly to costs for wastes from power-reactor fuels containing up to 1000-fold higher radionuclide concentrations.

Disposal as Solids in the Ocean Depths. Disposal of high-level wastes in deep ocean areas is a potential scheme that has received relatively little technical study. Basically such a scheme would involve converting the aqueous waste to a solid to permit transportation to the ocean location, transporting it to a deep part of the ocean floor (as deep as 36,000 ft), and anchoring it to the bottom, preferably under a layer of mud.[34] For radionuclides to reach man's environment, they would have to penetrate the solid waste itself, the waste container, the mud, and the deep ocean.

Simplified calculations[35] indicate that, if all the ^{90}Sr from only the U. S. nuclear power economy is evenly distributed in the ocean water and if the generation rate of nuclear power is assumed to double every 10 years beyond the year 2000 [with 940 MW(e)] to about 60,000 MW(e) in 2060, at that time the hypothetical perfectly mixed concentration of all the resultant ^{90}Sr in the ocean water would reach the present limits in 10 CFR 20 of *AEC Rules and Regulations*, or 3×10^{-7} μCi/cm^3.

The transport mechanisms for materials in the ocean waters are complex and not well known. For example, mixing times from some ocean depths to the surface are believed to be as long as several hundred to thousands of years, a beneficial factor for this application. On the other hand, possible concentration of some radionuclides in sea life which are then consumed by man requires reduction of allowable concentrations in seawater to below those stated in 10 CFR 20.

In view of the numerous unknowns in this concept and the international considerations, the scheme is not believed to be acceptable for use.

Disposal as Liquids in Deep Wells. The initial NAS committee reported "disposal of waste in porous beds interstratified with impermeable beds in a synclinical structure is a possibility for the more distant future."[25] Such a technique, termed deep-well disposal, involves injection of the liquid waste into permeable reservoirs in rock that are below and isolated from freshwater-bearing formations. The permeable formation, which normally contains naturally occurring fluids such as briny water, oil or gas, should generally be at least 3000 ft deep and overlain with layers of thick and impermeable formations such as shale or salt deposits.

The concept is based on a slow, lateral movement of fluid in the permeable formation, typically no more than about 3 ft per year. This movement, in combination with the width of suitable basins being tens to hundreds of miles in width, should provide containment in the formation for thousands of years.

Studies on the applicability of deep-well injection for disposal of highly radioactive wastes have been limited to engineering evaluations. The overall conclusions from these studies supported the original considerations,[25,36] with the premise that adequate proof be provided of solutions to potential problems for any specific application.[26,37] Studies would be required before application to confirm water flow rates and directions in the deep well, to confirm that there is complete lack of communication between the fluid in the well and water sources in our biosphere, to resolve the potential problem of plugging of the well with solids from the high-level wastes, to confirm the chemical compatibility of the aqueous wastes (which may require prior treatment) with the materials in the well, and to determine the effects of heat of decay on the performance of the system.

Because of the extensive and expensive studies required to apply deep-well disposal to highly radioactive wastes, because a reprocessing plant would have to be located over a suitable deep-well formation, and because of the progress made in studies on solidification and storage of wastes in salt mines, major experimental studies have not been performed and major development of this technique for high-level wastes is not being supported by the AEC.[27,37] However, deep-well disposal of the larger volumes of low- and intermediate-level radioactive wastes may become applicable since some of the potential problems with high-level wastes would not exist.

Perpetual Storage as Liquids in Deep, Underground Caverns. Storage of wastes as liquids or slurries in deep, underground caverns in stable, impermeable formations at the Savannah River site has been extensively investigated by workers at Savannah River, the U. S. Geological Survey, and the U. S. Army Corps of Engineers.[38,39] Although this concept was developed for long-cooled wastes with less radioactivity than high-level wastes as previously defined, it is presented here for information. The scheme has also been considered by workers at Richland for intermediate-level wastes.[19]

In this concept, shown schematically in Fig. 6, liquid waste is pumped through a pipe in a service shaft extending down to one of several lateral tunnels in a deep, thick, rock formation. When a tunnel is filled with waste, two thick concrete bulkheads are installed to seal off the tunnel from the central access tunnel. A number of other lateral tunnels would emanate from the central access tunnel as needed. The number, cross section, and spacing of the tunnels depend on the self-generating heat rate in the waste and the heat-transfer characteristics of the rock, so the pressure from boiling and gas formation within the tunnel will not violate the integrity of the surrounding rock. The proposed tunnel cross section for Savannah River waste is 18 by 30 ft.

Three primary barriers that tend to prevent migration of the radionuclides in this concept as proposed for the Savannah River site are the low permeability of the rock in which the vault is located (water movement at 1.5 to 7 ft/year), the thick impermeable layer of clay immediately above the rock formation, and the high ion-exchange capabilities of the sediments between the surface and the clay formation. It has been estimated that any one of these barriers is capable of confining the waste within the plant boundaries for at least 600 years. The integrity of the cavern is expected to be essentially unaffected by man-made disturbances at the surface and by earthquakes.[38,39]

Key requirements for the use of this scheme are the existence of suitable geology, hydrology, and other rock and soil characteristics at the reprocessing-plant site, because it is doubtful that transportation of aqueous wastes across private or public property will be permitted in the future. The time and expense of demonstration that these conditions exist for a commercial enterprise will severely deter the use of this concept. Also, the applicability of this scheme to high-level wastes has not been investigated.

Perpetual Storage as Liquids in Tanks. Storage as liquid wastes in tanks is not considered to be more
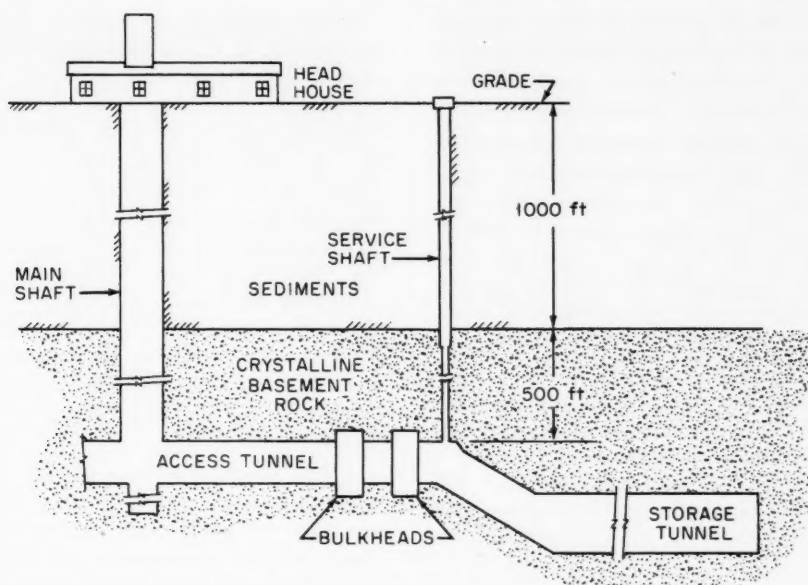
Fig. 6   Storage vault for radioactive wastes.

than an interim storage technique. Although no hazard to the public has resulted from this scheme to date and although the technology exists and is being used with a high degree of safety by this scheme, the potential consequences of failure could be large. Liquid storage is considered to be good for interim storage to permit decay of most of the short-lived radionuclides. When used for interim storage, the inventory of long-lived radionuclides in liquid waste (and thus the potential consequences of tank failure) is kept relatively small.

Perpetual storage by this scheme would require a high degree of positive controls over a nearly infinite time period. Such controls include continuous operating, monitoring, and surveillance, plus routine and continual replacement of each tank after 15 to 40 years of service. When a storage requirement of thousands of centuries is considered, such a scheme is most unrealistic.

Total costs[30] for perpetual liquid storage have been estimated by Blomeke to be approximately 0.03 to 0.035 mill/kW(e)-hr.

Ultimate Disposal

At this point in technology, no currently practicable schemes for unqualified ultimate disposal of radioactive wastes, as previously defined, have been developed, although storage in salt very nearly approaches ultimate disposal. At least two truly ulti-mate disposal schemes have been conceived, but neither is usable at this time and may never be. However, these or other concepts of this nature, which may evolve, may become practicable at some time in the future. The two concepts are presented here as examples of conceivable ultimate disposal.

Transmutation. The first concept[40,41] involves highly efficient separation of the long-lived radio-nuclides from the waste and then transmutation of the specific radionuclides in a spallation reactor to form short-lived radionuclides that decay to stable isotopes. The latter part of this scheme, conceived by Steinberg and others at Brookhaven National Laboratory for transmutation of ^{137}Cs and ^{90}Sr, consists of a nuclear reactor to provide the power to operate a high-energy proton accelerator that feeds a beam of protons into a spallation reactor where the transmutation takes place.

Although transformation of isotopes by this scheme is possible, the amount of mixed fission products generated would approximately equal the amount of ^{137}Cs and ^{90}Sr transmuted.[42] Also, the scheme does not provide for disposal of the important long-lived actinide radionuclides and the smaller amounts of long-lived fission products. In addition, the scheme would require costly and highly efficient separation of the ^{137}Cs and ^{90}Sr (and all other radionuclides for which separation is necessary) from the remaining waste, then conversion to a target for the

spallation reactor (in addition to the other "routine" waste-management costs of interim liquid storage until decay of short-lived radionuclides), and finally the transportation of the "target" to the spallation reactor.

Space Disposal. Another conceivable method for ultimate disposal of high-level waste is that of transporting the material away from our planet and into space, probably to a star. Primary present limitations to applying this disposal technique are the high costs and the reliability and safety of the launching and propulsion systems.

Assuming disposal of fission products, plutonium losses, and transplutonium isotopes only (i.e., fuel reprocessing is conducted without addition of nonvolatile process materials to the waste) as oxides, and the present cost capability of $1000 per payload pound,[43] the space transportation costs would be about 0.3 mill/kW(e)-hr. Using the projections of ultimate reductions in payload cost to $100 per payload pound,[43] the costs are reduced to about 0.03 mill/kW(e)-hr. Again, these costs do not include the other waste-management costs of processing to eliminate added impurities, interim storage of aqueous wastes, conversion of the fission products to compact solids, transportation to the launching site, remote handling at the launching site, and special safety precautions.

Transportation Considerations

The wastes from the fuel-reprocessing plant will have to be transported to the storage or disposal site, and technology exists to permit such transportation on a safe basis. However, transportation of wastes does add another step in the nuclear fuel cycle wherein careful considerations for safety must be applied.

The volume of solidified waste is in the same range as the gross volume of the original irradiated fuel elements, but the radioactivity level of the solid waste and its resultant heat generation rate is less by a factor of about 10, owing to the decay of radionuclides during the interim aging time. Therefore less shielding and less cooling are required for transporting solid wastes than for irradiated fuel elements. Furthermore, gaseous fission products such as ^{131}I and [85]Kr are not present in the solidified wastes. Consequently shipping problems are expected to be less for solid wastes than for irradiated fuel.

Shipment of high-activity waste will be governed by the *Code of Federal Regulations*[1,44] and the Department of Transportation (DOT) regulations. Engineering studies which have been carried out to date indicate that unacceptable safety and economic

problems would be encountered in the design and construction of a bulk-liquid high-level waste shipping system that would meet AEC and DOT shipping regulations. For example, the decay heat generated within the waste could result in the buildup of steam pressure within shipping casks following a loss-of-coolant incident; such conditions could, with a failure of containment integrity, lead to release of liquid waste to the environment. Also, radiolytic decomposition of the water to hydrogen and oxygen could result in a potentially explosive mixture in a few hours. Therefore it has been concluded that bulk quantities of liquid high-activity waste may constitute an unacceptable risk to public health and safety. Thus such wastes should be converted to a solid form prior to shipment beyond the fuel-reprocessing plant.[15]

The proposed regulations require that the waste be shipped in an approved solid form to a federal repository within 10 years after reprocessing of the fuel.[1] Approval of the various solid forms will likely be based upon the same safety criteria established for shipment of irradiated fuel materials.

Solidified wastes will likely be in the form of ceramic, glass, or calcine encased in a steel can sealed by welding. The cans will likely be cylinders (or some other equivalent shape) 6 to 24 in. in diameter and 8 to 10 ft long. The total shipping package (the solidified waste in its can, and the cask and its appurtenances) must satisfy the requirements of the regulations.

The most likely potential mechanism for release of radionuclides during transportation is from a fire or an impact, or a combination of both. In either case, the barriers preventing release of radionuclides are the solidified waste itself, the waste can, and the cask. Preliminary analyses indicate that, in a fire, the center line of the solidified waste could reach relatively high temperatures, but the walls of the can would remain at reasonable temperatures, probably no higher than about 400°C; thus the cask could withstand the accident. Furthermore, the pressure within the can would not be excessive, and the can would therefore remain intact.[30] Impact resistance of the cask will be heavily relied upon. However, even if the cask should fail owing to impact, the waste can is relatively rugged. Furthermore, characteristics of many types of solid wastes could provide additional safety assurance.

From the values in Table 1, the annual volume of solid wastes to be transported from the projected nuclear economy in the year 2000 (actual transport will take place between 2005 and 2010) is about 440,000 gal. Using casks with an average cavity diameter of 40 in., and shipping four 12-in.-diameter

waste cans containing 10-ft lengths of solid waste (7.8 ft³ each), approximately 1800 cask loads/year will be required. A cask of this size will weigh 50 to 75 tons. Present "routine" shipments by truck are limited to about 25 tons of total weight, with some occasional special permits for up to about 55 tons. Present rail limits are near 100 tons.

Costs for transportation of solidified wastes have been estimated by Blomeke[30] to be in the range of 0.001 to 0.003 mill/kW(e)-hr, or less than 10% of the total waste-management costs.

CONVERSION TO SOLIDS

The conversion of high-level liquid wastes to solids for long-term storage is being developed in nearly all countries with significant near-future nuclear energy as shown in Table 4. Solidification is being developed so widely because it is the only reasonably attainable technique for achieving a substantial increase in the safety associated with the storage and disposal of the high-level waste from the reprocessing of irradiated reactor fuel.

Overall Status of Solidification Processes

Much work has been done during the past 15 years, and is still in progress, for the development of techniques for solidification of high-level wastes. Four processes for solidification of these wastes have been developed in the United States to the point of radioactive demonstrations on an engineering scale. The four processes are pot calcination, spray solidification, phosphate-glass solidification, and fluidized-bed calcination.

Solidification of various types of high-level wastes has been demonstrated for the AEC using the pot, spray, and phosphate-glass processes on a full-level, engineering scale in the Waste Solidification Engineering Prototype (WSEP) by Battelle–Northwest at Richland, Wash., since November 1966. Demonstrations were to be completed in 1970. The fluidized-bed process has been demonstrated for the AEC in a large-capacity plant in the Waste Calcining Facility (WCF) by Idaho Nuclear Corp. at NRTS, operating on intermediate-level wastes since 1963. Most of the nonradioactive development work is completed, and most of the basic technology has been obtained.[12,45]

The pot, spray, and phosphate-glass processes have been developed and are being demonstrated at processing rates of 10 to 20 liters/hr of Purex liquid waste, or waste from about 1 metric ton/day of nuclear fuel.[12] The fluidized-bed process has been demonstrated at high rates of 300 liters/hr of liquid wastes containing large amounts of aluminum and zirconium salts. Scale-up of all processes for Purex wastes beyond about 1 metric ton/day equivalent fuel will require some modest additional nonradioactive development. An overall summary of the status of development of these and several other processes is presented in Table 4. Other processes have been developed to a lesser degree in this country, but discussion will be limited to the four processes with the furthest development since these four processes encompass most features of all process developments.

In all processes the waste is heated to 400 to 1200°C so that essentially all the volatile constituents (primarily water and oxides of nitrogen) are driven off and either a solid or a melt that will cool to a solid is left. The resulting solids are relatively stable chemically, especially at temperatures lower than those during processing.

General Chemical Considerations for Waste Solidification

The simplest chemistry for solidification is in the processes that form calcines. Calcines consist of mixtures of the nonvolatile oxides (and sometimes sulfates or fluorides, if present) of the metallic-salt constituents in the waste. All other material is volatilized. For calcines, the minimum weight of solidified waste (that of fission-product oxides alone) is about 1.1 kg/1000 MWd(t) exposure. Contributions from inert chemicals added during fuel reprocessing as shown in the compositions in Fig. 1 can increase that weight by a factor up to about 4.

Solidification processes that form melts require significant chemical modification of almost any waste composition. Compositions for waste solidification generally require at least 70 mole % of inert chemicals to incorporate the fission products into materials that are meltable at reasonably low temperatures, or at temperatures less than about 1000°C. (A more typical value for inert chemical content in melts is 80 to 85%.)

Melts have been developed in which the major melt-making fluxes are phosphates,[46-48] borophosphates,[49-51] silicates,[51,53] borosilicates,[49,52-54] and borates.[55] In most cases workable chemical-composition ranges for good solidified waste products have been defined. However, general correlations for chemical compositions are difficult to define because of the complex interaction of all the constituents in the wastes. Because of the complex chemistry, each waste composition encountered usually requires some laboratory investigation of melt-forming compositions.

Table 4   Summary of Research and Development on Solidification
of High-Level Waste

Process and sites*‡	Time span	Lab-scale radioactivity†	Pilot plant Radioactivity†	Pilot plant Capacity, liter/hr	Product	Chemical additives	Status of work
Pot calcination							
ORNL	1958 to 1965	None	None	25	Calcine	Calcium, sulfate	Done
BNW	1959 to 1962	None	None	10	Calcine	Sulfate	Done
	1962 to date	H	H	20	Calcine	Sulfate, calcium	In progress
Spray							
BNW	1959 to date	H	H	20	Ceramic, glass	Phosphate, Borophosphate	In progress
USSR	~1961 to date	?	?	20	Calcine glass	Borosilicate	In progress
Phosphate glass							
BNL	1960 to date	None	None	20	Glass	Phosphate	In progress
BNW	1964 to date	H	H	20	Glass	Phosphate	In progress
Fluid bed							
ANL	1955 to 1959	None	L	6	Granules	None	Done
INC	1955 to date	L	I	300	Granules	None	In progress
BNW	1959 to 1961	No work	None	20	Granules	None	Done
USSR	~1962 to date	?	?	30	Glass, granules	Borosilicate	In progress
Pot glass							
AERE	1959 to 1966	None	H	6	Glass	Borosilicate	Done
FAR	1962 to date	H	L	20	Glass	Boroalumino-silicate Phosphosilicate	In progress
CPP	1969 startup	No work	H	20	Glass	Boroalumino-silicate	In progress
ORNL	1961 to 1966	None	None	3	Semiglass	Phosphate Borophosphate	Done
Rotary kiln							
BNL	1955 to 1963	None	None	20	Powder	None	Done
FAR	1960 to date	None	None	6	Glass	Phosphosilicate Borosilicate	In progress
Ceramic sponge							
LASL	1959 to 1964	None	L	4	Ceramic balls	None	Done

*Work is also being done in Canada, Germany, Denmark, India, Japan, and Czechoslovakia.
†Values are based upon H is >70 Ci/kg of solid, I is 0.07 to 70 Ci/kg of solid, and L is <0.07 Ci/kg of solid.
‡Abbreviation summary:
  ORNL, Oak Ridge National Laboratory, Oak Ridge, Tenn.
  BNW, Battelle—Northwest, Richland, Wash.
  USSR, Union of Soviet Socialist Republics.
  BNL, Brookhaven National Laboratory, Upton, Long Island, N. Y.
  ANL, Argonne National Laboratory, Argonne, Ill.
  INC, Idaho Nuclear Corporation, Idaho Falls, Idaho.
  AERE, Atomic Energy Research Establishment, Harwell, Berks, England.
  FAR, Center for Nuclear Studies, Fontenay-aux-Roses, France.
  CPP, Center for Plutonium Production, Marcoule, France.
  LASL, Los Alamos Scientific Laboratory, Los Alamos, N. Mex.

Similar studies are usually necessary to predict occurrence of special problems, such as ruthenium or sulfate volatilization, foaming, and stickiness, for all solidification processes. The presence of the cations of iron, sodium, and aluminum in the aqueous wastes usually enhances the formation of meltable compositions.[12]

Three of the inert chemical constituents that may be in high-level wastes are sufficiently troublesome in solidification processing to merit effort to keep them out of high-level wastes. These constituents are sulfate, fluoride, and mercury ions.

The sulfate ion is generally unstable chemically at the higher range of temperatures reached in solidification (700°C and higher) and tends to volatilize. Retention of sulfate in the solidified waste at temperatures above 700°C requires chemical additives (usually calcium). Retention above 850 to 900°C is very difficult, particularly in melts.[46] This volatilization can result in added corrosion problems in the off-gas system, recycle of sulfates, and increased sulfate concentrations in the liquid waste for cases of partial volatilization or can produce another medium-to-high-level waste stream that requires special treatment and disposal for cases of complete volatilization. Sulfate can also cause severe precipitation and resultant solution-handling problems, as discussed previously.[11,56]

Fluoride is retained with difficulty (by using calcium) in solidified-waste processing up to temperatures of about 600°C.[57] At higher temperatures a significant fraction of fluoride may volatilize and have to be recycled or disposed of by other means.[58] Fluoride content of much more than about $0.001M$ will significantly increase corrosion of stainless steel and titanium (used generally in waste-processing systems), although this corrosion can be largely overcome by use of complexing agents (aluminum, zirconium, etc.).

Mercury cannot be retained in the solidified waste when processed at temperatures above 400 to 500°C. When volatilized, the mercury and its oxides condense at temperatures of about 350°C and present relatively serious potential plugging problems. A means for pretreating the aqueous waste for removal of mercury has been developed in the laboratory.[59]

Ruthenium, one of the ever-present fission products, is just as cantankerous in waste solidification as it is in fuel reprocessing. One to 80% of the ruthenium will usually oxidize and volatilize during solidification, and it is more difficult to remove from the off-gas stream than are the entrained nonvolatile materials. Chemical additions to minimize the oxidizing potential during solidification are sometimes required to mini-

mize oxidation to the volatile RuO_4 form. Even then, volatilization of at least 1% is usually encountered. Volatilized ruthenium can be recycled to the solidification step or removed by filters and selective chemical treatment devices.

All processes for solidification of high-level waste generate additional waste streams that contain intermediate levels of radioactivity. These are the vapor or condensate streams from the solidifier which have radionuclide contents less than the aqueous high-level wastes by factors of 10 to 1000. These primary effluents from solidification must be decontaminated by additional factors up to 1×10^{10}, which is comparable to those of the system for handling high-level liquid waste in a fuel-reprocessing plant. Processing of these vapor streams would logically be done by recycle routing to the existing high-level liquid-waste concentration and processing equipment. Only a modest increase in capacity (on the order of 10%) of the liquid-waste processing capacity of the reprocessing plant would be required.

A solidification plant could not be located remote from the fuel-reprocessing plant because violation of proposed policy would be required to ship the high-level liquid waste to the solidification plant and the recovered nitric acid with low-to-intermediate-level radioactivity content back to the separations plant.

Solidification Processes

The four principal processes—pot calcination, spray solidification, phosphate-glass solidification, and fluidized-bed solidification—are shown in Fig. 7.

Pot Calcination. Pot calcination, which was developed at ORNL, is a batch process that has been developed to a state of readiness for commercial radioactive use.[60] Pot calcination has the advantage of being a simple process that is adaptable to a wide variety of feed compositions. Its disadvantages are that a stainless-steel pot is required, the thermal conductivity of the calcine is low, the capacity of a system must be increased by multiple pot lines, and the solidified waste is quite leachable in water.

In pot calcination the principal processing vessel, the pot, is also the final container for the solidified waste. In this process (shown in Fig. 7a), liquid waste is added to a pot that is heated in a multiple-zone heating and cooling furnace. The waste is concentrated at a constant volume to the point that a scale of calcine cake forms on the walls of the pot. As calcination continues, the scale grows in thickness and reduces the
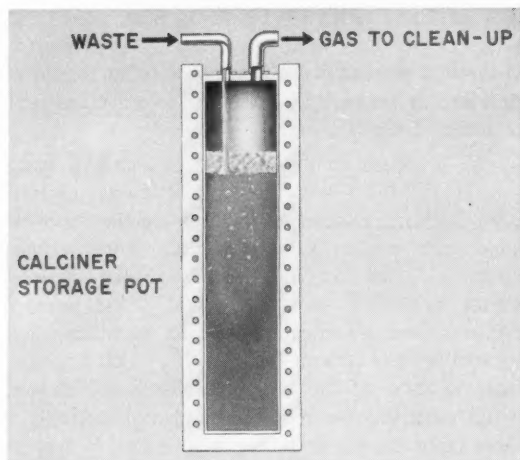
(a) Pot calcination.
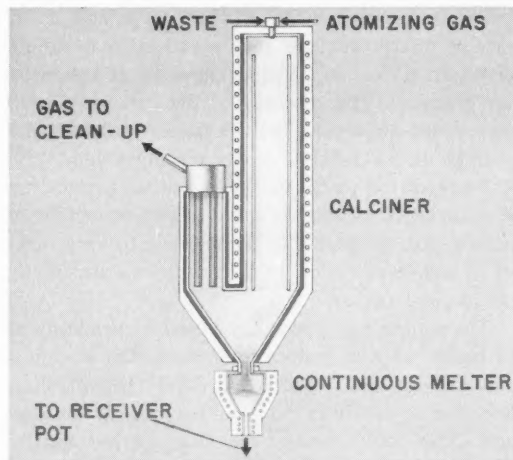
(b) Spray solidification.
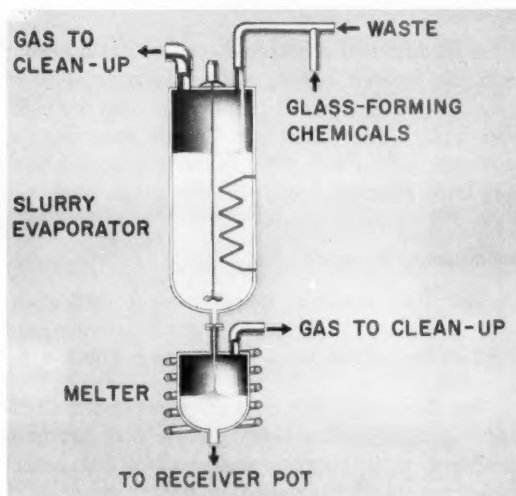
(c) Phosphate-glass solidification.
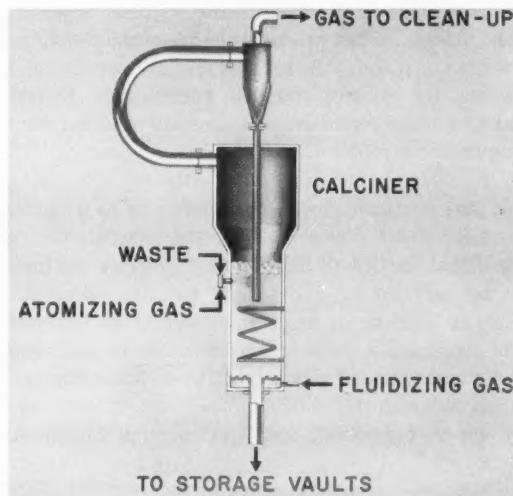
(d) Fluidized-bed calcination.

Fig. 7 Primary solidification techniques in the United States.

heat transfer from the pot wall to the boiling sludge, and therefore the feed rate must be reduced proportionately. When the feed rate is reduced to an unprofitable rate (about 5 liters/hr), the feed is shut off. At this point the scale has grown inward from the pot wall and upward from the bottom of the pot to fill the pot, except for a thin-cone-shaped liquid-containing space in the upper 2 to 3 ft of the salt cake.

Heating is then continued until the liquid is boiled to dryness and all the waste in the pot has been calcined and has reached the temperature of 850 to 900°C where essentially all of the volatile constituents have been released. The pot is then cooled in the furnace, removed, sealed, and taken to storage.

Because the pots serve as the processing vessels, they must be made of corrosion-resistant material to

withstand the severe corrosion conditions during calcination. Corrosion of 304L stainless steel is negligible during processing (less than 0.0003 in./day). Pots must have liquid-level and temperature-measurement devices. Liquid level may be measured either with a standard gas-purged dip tube or with an internal temperature sensor near the top of the pot. In demonstration tests, temperature measurements are taken at the center line and at the pot walls in each zone with in-place thermocouples.[4,11,12,60] Because of the significant cost of these thermocouples, an incentive exists in commercial use for reducing the number of thermocouples or for making the thermocouples reusable.

Internal heat from decay of radioactive constituents requires slight modifications of operating techniques. With the presence of internal heat, cooling of the pot wall is necessary before the material at the center of the pot has reached its final maximum temperature or the center temperature will exceed that desired. (Higher temperatures can result in severe corrosion and undesirable volatilization of some constituents.) Control of this temperature has been successfully demonstrated by using a simple three-step reduction of furnace temperatures, based on pot-wall and -center temperatures.[11]

The pot calcination cycle may be divided into three major periods: (1) feeding and concentrating at constant feed rate, (2) pot-wall scaling and calcining, causing gradual reduction of feed rates, and (3) calcining and cooling, when feed is turned off and the calcine is heated to 850 to 900°C and then cooled in preparation for removing the pot from the furnace. Typical time requirements for each of the steps are summarized in Table 5. Since pot diameter has a relatively small effect on the overall processing capacity, an increase in capacity must be obtained by changes in pot geometry (e.g., annular pots) or by multiple pot lines.

Longer pots provide a slight increase in capacity because there is no marked increase in calcining and cooling times. Maximum boil-up rate is limited by entrainment in the upper part of the pot or by the cross-sectional area of the pot. Some wastes may contain significant amounts of foam-making constituents (e.g., dibutyl phosphate from the reprocessing plant). If foaming is present, feed rates during the initial boiling period must be reduced from those shown in Table 5.

During pot calcination of Purex wastes, ruthenium is volatilized to the extent of about 5% and 10 to 30% for low-sulfate wastes and high-sulfate wastes, respec-

Table 5 Time Cycle and Capacities for Pot Calcination*†

Pot diameter, in.	8	12
Pot height of fill, ft	6	6
Volume of calcine, liters	60	120
Feed volume, liters	500	1000
Initial feed rate, liters/hr‡	30	60
Time cycle		
Time of initial feed rate, hr‡	10	10
Time of reducing feed rate, hr	20	30
Calcining and cooling time after feed is turned off, hr	10	30§
Total time cycle, hr	40	70
Overall cycle capacity, liters/hr	12	14
Equivalent waste processing capacity, metric ton/day with feed concentration = 378 liters/ metric ton	0.75	0.9

*From Ref. 11.
†For pots containing 5 kW of internal heat.
‡For feeds relatively free of foaming tendencies.
§Estimated; exact data not available.

tively, although other variables may also have affected volatilization.[11] Lower volatilization can be effected by the addition of chemical reductants, such as nitric oxide gas or phosphites.[61]

Sulfate is not normally present, but, if it is present in the waste, less than 2% of the sulfate will be volatilized from the calcine if the chemical composition of the waste is adjusted to assure that the chemical sulfate equivalent of alkali- or alkaline-earth metallic ions is present. In practice, sodium and/or calcium nitrates are usually used.

Volatilization of cesium and rubidium, which are always present as fission products, can be virtually eliminated by adding enough sulfate or phosphate ions to the feed to be chemically equivalent to the total amount of alkali metals present.

Entrainment from the pot calciner, when operated on a reasonably conservative capacity basis, is approximately 0.4% of the total feed.[11]

Recent laboratory studies with borosilicate-glass compositions are expected to allow the process to be operated as a rising-level glass-making process. Compositions and time at operating temperature can be adjusted to provide a dispersion of waste in a glassy matrix with this scheme.[62] Radioactive demonstrations of this method were planned for 1970.

Spray Solidification. Spray solidification is a continuous process that has been extensively developed

and is approaching readiness for commercial use.[12,14] The spray-solidification process, developed by Battelle—Northwest, has several advantages. It is a continuous process with low holdup volumes, it is adaptable to a moderately wide variety of feed compositions, and it produces a variety of good-quality solids. Its disadvantages are that it is a moderately complicated system, it requires good flow control of sometimes difficult-to-handle feed solutions, its performance requires high-quality atomization, and at present it requires the use of a relatively expensive platinum melter. Recent pilot-plant testing of melting the calcined powder in the receiver pot, rather than using an expensive platinum melter, has eliminated one of the disadvantages.

In the spray calciner (see Fig. 7b), liquid waste (which contains some or all of the melt-making additives) is fed through a pneumatic atomizing nozzle into the top of a heated cylindrical tower. The atomized waste is sequentially evaporated, dried, and calcined to a powder as it falls into a continuous melter below the calciner tower where it is melted at temperatures of 800 to 1200°C. Process gases from calcination flow into the adjacent filter chamber, carrying much of the calcined powder as dust. The dust collects on the porous metal filters as the gas passes through. The dust deposits are periodically blown off the filters by sudden pulses of high-pressure steam or air directed backward through the filters by small nozzles. The dislodged dust falls into the melter with the main powder stream. The molten calcine flows through an overflow weir or a freeze valve into the receiver—storage pot below. After the pot is filled, it is cooled in the furnace, sealed, and taken to storage.

In the continuous melter the small amount of residual nitrate and water is driven off from the calcine, and the calcine is melted. Platinum is the only reliable metallic construction material found to date to withstand the corrosive environment and high temperature with phosphate melts, although other materials appear promising. Platinum has been used extensively at temperatures up to 1250°C. A special alloy of 50% Cr—50% Ni is generally satisfactory at temperatures up to 1000°C and a chromium—alumina cermet apparently is satisfactory at temperatures up to 1100°C. High Cr—Ni stainless steel and some high-nickel alloys are satisfactory at temperatures up to 900°C.

The capacity of a platinum melter that is 10 in. in diameter and has a 14-in.-high heated section is 1.7 liters/hr of melt.[55] Discharge of melt from the melter has been adequately demonstrated on a continuous basis using overflow weirs or batchwise using straight-tube freeze valves wherein a plug of melt about 2 in. long is melted or frozen to provide on—off flow control.

The pot for receiving the molten waste may be made of mild steel if the pot is filled with melt by large, rapid, batchwise dumps from the melter or if the pot is filled with melts with low melting points (less than about 700°C). These limitations are caused by the need to heat the pots under slow-filling conditions to the point where the melt will slump to assure complete filling of the pots without formation of stalagmites or voids. (Mild-steel pots can acceptably resist oxidation by air at temperatures up to about 650°C for periods of several days.) Corrosion of mild-steel or stainless-steel pots by phosphate melts at temperatures about 700°C or lower is negligible.[4,63] Provisions for the batch filling of pots from this and other continuous solidification processes must be accommodated in the design.

Most of the flow sheets used in spray-solidification processing at Battelle—Northwest produce alkali metal—phosphate solids. These compositions are used primarily because they offer relatively large latitude in chemical composition, they have generally low melting points (700 to 900°C), they produce melts with reasonably low viscosities (less than 50 P) at operating temperatures, the chemically adjusted feed solutions are relatively easy to handle, they generally produce homogenous melts, and the final solid has desirable characteristics. The primary disadvantages of phosphate melts are those of higher corrosion rates and somewhat higher leach rates than other melts, such as silicates and borosilicates. With the typical phosphate melts, microcrystalline solids are formed in spray solidification by enough phosphate addition to approach orthophosphate melts (Σ normality of cations/phosphorus = 2.5 to 3.0). Sufficient alkali metals are added to reduce the melting point to 700 to 900°C.

Sulfate is retained in melts when calcium is added in excess to combine chemically with the sulfate. In some cases a small amount of aluminum is added to increase the sintering temperature for better operation in the spray calciner. Some of the chemicals are added as solids directly to the melter to permit operation of the spray calciner with a chemical composition that has a higher melting point than that of the final melt. With conditions used in the spray solidifier, approximately 95% of the sulfate is retained in the final solid.

Up to 75% of the ruthenium can be volatilized during the spray-calcination step (not during melting) with the phosphate flow sheets.[14,64] This volatiliza-

tion can be reduced by completely eliminating melt-making flux from the feed and addition of all melt-making flux to the melter, by reducing the oxidizing potential in the calciner, by reducing the nitrate and acid concentrations in the waste, and by using borosilicate fluxes.[14,65] Volatilization of the fission products cesium and rubidium has not occurred significantly in spray solidification flow sheets.

Recent studies have shown the applicability of two modifications to the conventional spray-solidification process: (1) the use of "in-pot melting" techniques wherein the melter is eliminated and additional melt-making solid flux is added directly to a stainless-steel pot where the melting is done,[65] and (2) the use of borosilicate fluxes for melting (with a melter, or with in-pot melting).[54] Radioactive demonstrations of these modifications were completed early in 1970.

Phosphate-Glass Solidification.   Phosphate-glass solidification has been extensively developed at Brookhaven National Laboratory (BNL) and can soon be put to commercial use.[63,66] It has the advantage of being a continuous process that produces a good-quality glass product. Its disadvantages are that it is a moderately complicated system, it requires operation with slurries that are difficult to handle, it cannot retain sulfate in the final solid, and at present it requires the use of a relatively expensive platinum melter.

Liquid waste containing all the melt-making additives is first fed to the evaporator where it is concentrated and denitrated by factors of 2 to 10 to a thick, syrupy, aqueous phosphate slurry.[67] The slurry is fed to the continuous melter where the remaining water, nitrates, and other volatile constituents are volatilized, and the material is heated to 1000 to 1200°C to form a molten glass. The molten glass flows through an overflow weir or a freeze valve into the receiver—storage pot below. After the pot is filled, it is cooled in the furnace, sealed, and taken to storage.

The net heat-transfer requirements for the phosphate-glass melter are somewhat higher than for the spray-solidifier melter, primarily because of the added evaporation load. In addition, the desired freeboard requirements above the melt level are somewhat higher because of the foaming tendency in the melter.[68] A platinum melter is 10 in. in diameter and has a 14-in.-high heated section with a capacity of 1.4 liters/hr of glass, or about 4 liters/hr of slurry feed.[13,69] The vapor stream from the melter is hot (400 to 600°C) and corrosive[69,70] and must be routed through platinum piping until the temperature is reduced to about 120°C.

The pot for receiving the molten glass is similar to that for the spray-solidification process. The lower slump point (600 to 700°C) and the continuous viscosity—temperature relation for the phosphate glasses permit filling of pots by slow continuous dripping of melt while the pot is heated to only 500 to 600°C. The corrosion rate on mild steel under these conditions is tolerable. Provisions for the batch filling of pots from this and other continuous solidification processes must be accommodated in the design.

Solidification of high-level waste solutions that contain sulfate can readily be done using the phosphate-glass process, but the sulfate is completely volatilized from the melter. In this case the vapor stream from the melter forms a separate stream of intermediate-level waste. This stream contains all the sulfate and normally about 30% of the nitrate, 2 to 10% of the radioruthenium, and less than 0.3% of all other radioactivity that was originally in the liquid-waste stream.[65] Because the sulfate cannot be reused, it requires special treatment for final disposal. When sulfate is not present, the condensate from the melter contains only nitrates and can be recycled with the condensate from the denitrator—evaporator to reduce the radionuclides in the off-gases from the solidifier to less than about 1% for radioruthenium and 0.5% or less for all other radionuclides.

The chemical adjustments required for the phosphate-glass process consist mainly of adding phosphoric acid to the feed to obtain a metaphosphate melt (Σ normality of metal ions/phosphorus = 1). The molar ratio of the oxides of the alkali metals is maintained at about half that of the total metal oxides in the melt to obtain a glass that forms at a reasonable temperature (850 to 1000°C), remelts at a low temperature (650 to 700°C), and has good handling properties. The chemically adjusted feed to the denitrator—evaporator is usually moderately friendly——the solids are gelatinous and are readily suspended. Concentration in the denitrator—evaporator sometimes progresses through stages of foaming or heavy crystalline deposits at lower and higher than normal concentration factors.[13,67] These conditions must be defined for each flow sheet.

Fluidized-Bed Solidification.   Fluidized-bed solidification is a continuous process that has been extensively developed for use with aluminum nitrate and zirconium fluoride—aluminum nitrate wastes. Development of fluidized-bed solidification was initiated at Argonne National Laboratory and has been extensively developed and demonstrated by Idaho Nuclear Corp. at NRTS. The process is ready for commercial application

for the above-mentioned wastes at NRTS, but development with the more unfriendly Purex wastes has been limited. The advantages of the fluidized-bed process are that it is continuous, it has a relatively high capacity for a given equipment size, its scale-up technology is relatively well known, and its solidified waste products are readily transportable by pneumatic means. The disadvantages of the process are that it is a moderately complicated system and its solidified waste products are relatively water soluble and have relatively poor thermal conductivities.

In fluidized-bed solidification the liquid waste is injected through pneumatic atomizing nozzles into the side of a heated (400 to 600°C) bed of granular solids which is continuously agitated (fluidized) by gas sparging upward through the fluidized-bed reactor. Contact of the waste with the hot, granular bed results in evaporation and calcination of the feed as coatings on the bed particles. The calcine entrained with the process gases from the calciner is removed from the gas stream by cyclone separators or filters and is then returned to the main stream of particles.[71] The main stream of particles is continuously removed from the reactor and transported to storage tanks.

For small fluidized-bed calciners (less than about 12 in. in diameter), the heat has usually been provided solely through the walls of the reactor, using conventional electric heating systems.[72,73] For larger reactors (and for some smaller reactors), additional heat has been added through heat-transfer surfaces inside the reactor bed (and by internal exothermic chemical reactions) to provide better heat distribution. Internal heat must be provided with controlled maximum temperatures to prevent localized formation of hot spots and caking.[72,74,75] NaK has been demonstrated as a heat-transfer fluid, and combustion of fuel oil is now being used for heating at the WCF at NRTS.[18,33] For high-level wastes with high rates of self-heat generation, the fluidized-bed system requires a means for cooling the contents of the bed or for dumping the bed during shutdown periods. Such provisions will eliminate the potential for self-overheating of the bed when flow of feed to the bed has stopped.

Containers for fluidized-bed calcine may be either the individual pots discussed previously or the large cylindrical or annular containers demonstrated at the WCF.[33,76,77] Storage containers for fluidized-bed calcine have been made of stainless steel, although the use of mild steel may be possible. Provisions for the batch filling of pots from this and other continuous solidification processes must be accommodated in the design.

The relatively limited development that has been done on Purex wastes indicates that successful calcination of the Purex wastes by fluidized-bed calcination is most likely. Purex wastes are less friendly than aluminum wastes because of their greater solubility in the feed solution and the low melting point and relatively high temperature of decomposition of the sodium nitrate in the wastes. Although these characteristics tend to cause increased agglomeration of particles and increased formation of lumps around the nozzle, control of agglomerate formation by impingement air-jet grinding and variations in fluidizing gas rates and by simple modifications to commercial atomizing nozzles appears to solve these problems.[73,74] General Electric Company's Midwest Fuel Reprocessing Plant at Morris, Ill., which is expected to be in full radioactive operation in 1971, will be using fluidized-bed calcination of Purex wastes by employing modifications of the calcination technique.[78]

Ruthenium volatilization from aluminum nitrate wastes[72] varies from less than 1% at 550°C to greater than 90% at 350°C, with an average volatilized amount of 40% in the WCF operating[76] at 400°C. Addition of chemical reductants greatly reduces the volatility of ruthenium. Volatilization from Purex wastes at 500°C was about 70% and was reduced to about 1% when sugar (a chemical reductant) was added to the feed.[73]

Greater than 99% of the sulfate is retained with the calcine in fluidized-bed calcination of Purex waste.[73,74] Fluoride is retained with the calcine (99%) and corrosion is controlled in fluidized-bed calcination of zirconium fluoride—aluminum nitrate wastes by the chemical addition of calcium in stoichiometric equivalence to the amount of fluoride present.[57]

Characteristics of Solidified Wastes

The main incentive for converting high-level radioactive liquid wastes to solids is for improved containment and safety before or during the centuries of time the material must be stored before the radioactivity has decayed to nontoxic levels. The most important concern is during the first 10 years after fuel reprocessing when the heat-removal problems, temperature, radioactivity, and rate of temperature change are the greatest, and when the wastes are transported to the disposal or long-term storage site.

Desired Characteristics. To maximize the increased safety resulting from conversion of wastes to solids for storage, the desired characteristics of solidified wastes with primary importance are:

- Good thermal conductivity.

- Low leachability.
- Good chemical and radiation stability.
- Mechanical ruggedness.
- Noncorrosiveness to container.
- Minimum volume.
- Minimum cost.

The net effect of high thermal conductivity is to increase the amount of allowable heat stored in a pot. This characteristic permits reduction in the amount of time liquid waste must be stored before solidification and may permit decreases in the volumes of solidified wastes. These effects are summarized in Fig. 8 for wastes from light-water reactors at a waste volume of 1 ft^3/metric ton of fuel. The heat content of wastes from future fast reactors can be several times those in Fig. 8 for the first few years.

Low leachability of the solidified products is desired to minimize the amount of radioactivity that could be released in the event water contacts a breached container of solidified wastes. The safety of solidified products with high leach rates may depend

more on containers with higher degrees of integrity than products with lower leach rates.

Generally, leach rate of a melt-solidified waste decreases as the solidification temperature is increased and decreases with increased glassiness of the material.[12,47,48] Devitrification of many glasses into microcrystalline materials can and does occur with extended periods at high storage temperatures and with slow cooling.[48,79-83] (It is therefore possible that glasses will devitrify under "normal" processing and storage conditions.) When devitrification occurs, leachability is increased by about one order of magnitude.[4,12,82] On first contact with water, leachability of melt-solidified waste is relatively high; then over a period of 10 to 50 days it decreases by about a factor of 10 to a relatively steady rate.[4,12,81,83]

Chemical stability and resistance to radiation of solidified waste are important, primarily, to assure that gases which may significantly affect the integrity of the product or container are not generated during storage and, secondarily, to assure that the basic structure and
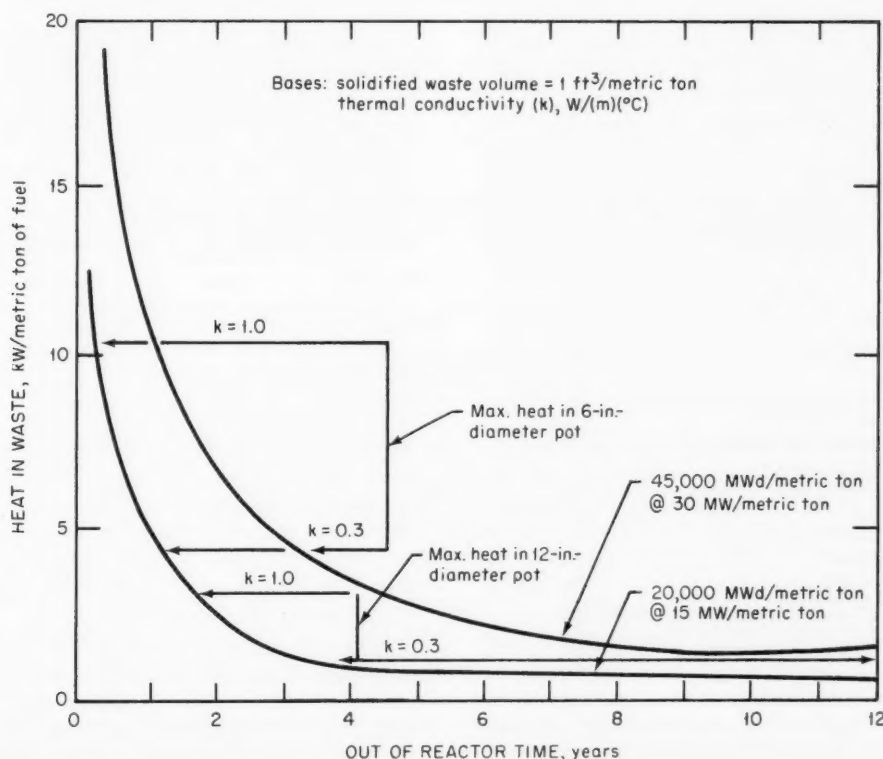
Fig. 8 Effects on heat removal from solidified-waste containers.

properties of the solidified waste are known. Experience to date indicates that gas formation from solidified waste in enclosed containers is generally not significant if storage temperature does not approach processing temperature.[4,11-14,76,80,81,84] At storage temperatures above processing temperatures, volatilization of some constituents has been found. For example, at 800°C, significant volatilization of cesium and ruthenium occurs from alumina solids made from the fluidized-bed process;[76] at 1200°C or higher, boron is volatilized from borosilicate glasses, and some phosphate is volatilized from phosphate melts.[12,81]

The eventual decay of radioactive elements to other chemical elements can represent up to 10% of the oxides present in calcined wastes and up to 5% of the oxides present in melt-solidified wastes. The effects of these changes on properties of the solid wastes are not well defined. Other characteristics may also change with time. For example, some glasses will devitrify to microcrystalline structures if held at 400 to 800°C for days or weeks,[4,12,48,77,83] calcined alumina granules change from amorphous to crystalline form, some volatile constituents migrate from thermally hot locations and condense at cooler locations, and phosphates and other glasses sometimes exude liquids.[4,76,77,79]

Mechanical ruggedness of the solidified-waste package is desirable primarily during transportation. In the event the container is breached, the ruggedness of the solidified waste is important in terms of its tendency to be dispersed. For example, in event of an impact accident, waste that has moderate leachability but is physically tough may result in less contamination to the environs than a waste that has low leachability and is brittle.

Corrosiveness of the solidified waste determines, in part, the life of its container. Data obtained thus far have indicated no significant corrosion rates in limited measurements,[4,11-14,60] but very-long-term effects have not been evaluated. The useful life of the containers is not known at this time but is expected to be much longer than the 15 to 40 years for containers for liquid wastes. In fact, data indicate that the life of mild steel or stainless steel stored in air will be determined by the oxidation rate from air on the outside of the containers.[4]

Minimum volume is desirable primarily for economic reasons. In general, reducing the volumes will reduce the size and cost of containers, of container storage areas, of shipping equipment, and of land for storage areas. Minimizing cost, without affecting quality, is an obvious advantage.

Actual Characteristics. Shown in Table 6 are the general characteristics of the solidified wastes from the four processes. Some of the characteristics are also illustrated by a photograph of samples in Fig. 9.

Table 6   Characteristics of Solidified High-Level Purex Wastes

	Pot calcine	Spray melt	Phosphate glass	Fluid bed
Form	Calcine cake	Monolithic	Monolithic	Granular
Description	Scale	Microcrystalline	Glass	Amorphous
Fission-product oxides, mole %	Up to 80	Up to 20	Up to 25	Up to 50
Bulk density, g/ml	1.1 to 1.5	2.7 to 3.5	2.7 to 3.0	1.0 to 1.7
Thermal conductivity, Btu/(hr)(ft)(°F)	0.15 to 0.25	0.5 to 1.0	0.4 to 1.0	0.10 to 0.25
Maximum heat, W/liter of solid*	85	220	190	70
Leachability in cold water, g/(cm^2)(day)	1.0 to 10^{-1}	10^{-3} to 10^{-6}	10^{-4} to 10^{-7}	1.0 to 10^{-11}
Hardness	Soft	Hard	Very hard	Moderate
Friability	Crumbly	Tough	Brittle	Moderate
Residual nitrate, wt.%	≤0.05	≤0.005	≤0.005	≤4.0
Volume, liters/1000 MWd(t)	1 to 2.5	1.2 to 3	1.5 to 5	1.5 to 5
Maximum stable temperature, °C	~900	Phase separation at ~900	Devitrifies at ~500	~600
Container material	Stainless steel	Mild steel or stainless steel	Mild steel or stainless steel	Mild steel or stainless steel

*Approximate values for storage in air in 8-in.-diameter cylindrical pots to maintain pot centerline temperatures less than 900°C and pot-wall temperatures less than 425°C. Average k values were used.

Spray solidification.
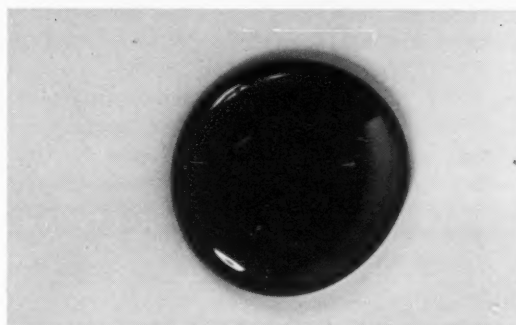
Phosphate glass.
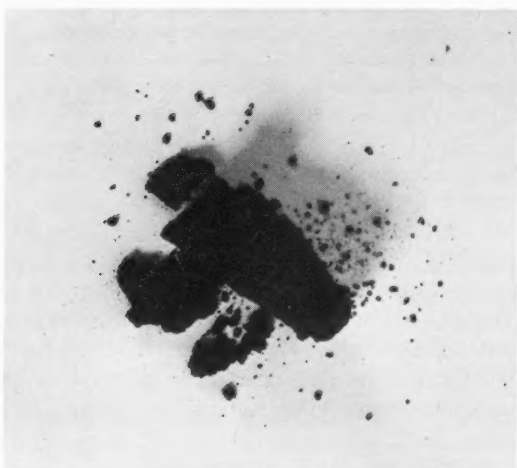
Pot calcination.

Fluidized-bed calcination.

Fig. 9 Samples of solidified high-level waste.

The solid waste from pot calcination is an oxide cake. It has a moderate bulk density with a void fraction of about 50%, has a low thermal conductivity, has relatively high leachability in water, and is relatively soft and friable.

The solid waste from spray solidification is a monolithic, rock-like, microcrystalline material. It is relatively dense with a low void fraction of up to 10%, has good thermal conductivity, has low leachability in water, and is hard and tough..

The solid waste from phosphate-glass solidification is a monolithic glass. It is relatively dense with a low void fraction of up to 5%, has good thermal conductivity, is very hard but brittle.

The solid waste from fluidized-bed calcination is a granular free-flowing calcine. It has a moderate bulk density with a high void fraction on the order of 50%, has low thermal conductivity, has relatively high leachability in water, and is moderately soft and friable.

An interesting comparison for leach rates shown in Table 6 is that the leach rate of Pyrex glass is approximately 5×10^{-7} g/(cm^2)(day).

To minimize the cost of high-level-waste management, the fuel reprocessor will package his high-level solidified waste in the largest container consistent with heat dissipation, safety, and transportation considerations. The resultant packages of waste will generally be

in the size range of cylinders 6 to 24 in. in diameter (or the equivalent size in other shapes). Shown in Fig. 10 are 8- and 12-in.-diameter (8 ft long) containers used in the WSEP.[70] Containers used in commercial applications will probably be similar but will probably not require as many auxiliary features (e.g., the large number of temperature measurements and the specially machined heads) as the developmental WSEP program. An extensive program is well under way in the WSEP to evaluate short-to-medium term changes in characteristics of the solid waste.[12,85]

Fig. 10 Containers for solidified waste in the waste-solidification demonstration program.

Table 7   Cost Range for Management of High-Level Waste from LWR Fuels*

	Probable time range, years	Cost for time range, 10^{-3} mill/kW(e)-hr	
		Min.	Max.
Interim liquid storage	For 0 to 5		22
Pot calcination†	At 0 to 5	8	22
Interim solid storage	For 0 to 10		10
Transportation of solids	At 3 to 10	1	3
Burial in salt	At 5 to 10	3	11
Comparison of total costs for specific cases			
Management of high-level wastes		37	45
Perpetual storage of liquid wastes		30	35

*33,000 MWd/metric ton of exposure.
†Range for a volume of 1.0 ft^3/10,000 MWd(t).

ESTIMATED COSTS

A number of cost-estimate studies have been made by Blomeke and coworkers at ORNL.[30] These studies have investigated the total costs of managing high-level wastes, which include interim liquid storage, conversion to solids by pot calcination, interim solids storage at the fuel-reprocessing plant, transportation of solids to long-term storage in a salt mine, and handling and burial of the solid waste in a salt mine. Partial costs of other schemes for storage or disposal of wastes were discussed earlier.

Costs of conversion to solids by processes other than pot calcination have not been determined in detail, but preliminary estimates indicate that conversion costs for the spray and phosphate glass processes may be somewhat higher than for pot calcination.[86] The costs of the other aspects of waste management for these two solids should be similar to those for pot calcine. Estimates of costs for converting power-reactor wastes to solids by fluidized-bed calcination have not been made, but the costs should be comparable to those for pot calcines.

A summary of the cost studies by Blomeke et al. is presented in Table 7. For the bases and cases used,‡ costs were 0.037 to 0.045 mill/kW(e)-hr, or $9500 to $14,700 per metric ton of fuel processed. Comparable costs for perpetual storage as liquids vary from 0.030 to 0.035 mill/kW(e)-hr, or near the lower end of the range for storage as solids.

From these studies the cost range for waste management by conversion to solids represents an increase of approximately 0.007 to a maximum of 0.02 mill/kW(e)-hr above that of perpetual liquid storage. These increases represent approximately 0.2 to 0.5% of the total cost of nuclear electric power. The cost of total waste management by any of the schemes represents approximately 1% of the total cost of nuclear electric power, or up to 20% of the total cost of reprocessing irradiated fuels.

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‡Reprocessing plants with capacities of about 6 metric tons/day; LWR fuels exposed to 33,000 MWd/metric ton at 30 MW/metric ton; 20-year plant life with straight-line depreciation; cost for private financing: 16% return on equity, 30% capital investment financed by bonds, 8% bond interest, escrow fund established with 5% interest, 48% federal income tax rate; pot calcination in cylindrical pots 6 to 24 in. in diameter to restrict maximum calcine centerline temperature to 900°C; 1.0 to 1.5 ft^3 of solid calcine for each 10,000 MWd(t); 2000-mile round-trip shipment in casks weighing 50 to 90 tons; long-term storage in salt mines.

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